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CONTROL TECHNIQUES FOR MERCURY EMISSIONS FROM EXTRACTION AND CHLOR-ALKALI PLANTS

Environmental Protection Agency
Research Triangle Park, North Carolina

February 1973

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The report deals with two sources of mercury emissions, the primary mercury processing industry and the mercury-cell chlor-alkali industry. An effort is made (1) to identify the process steps that may produce atmospheric mercury emissions, (2) summarize the emission control technique and low mercury emission processes used or applicable to these industries, and (3) to evaluate these techniques in terms of cost and effectiveness. The condenser gas stream is the major source of mercury emissions from a primary mercury processing plant. The amount of emissions can be reduced by converting to processes that inherently produce fewer emissions or by treating effluent gases to remove mercury. Appropriate control techniques include cooling and mist elimination, wet scrubbing, or adsorption beds. Major emissions of mercury from a chlor-alkali plant using mercury cells are from the hydrogen gas stream, the end-box ventilation stream, and the cell room ventilation air. The emissions from all sources can be eliminated by converting to the diaphragm-cell process. The cost of converting a 100-ton-per-day plant is estimated to range from $3,700,000 to $8,000,000.

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ENVIRONMENTAL PROTECTION AGENCY
Office of Air and Water Programs
Office of Air Quality Planning and Standards
Research Triangle Park, North Carolina 27711
February 1973

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PREFACE

This document contains information about the nature and control of a hazardous air pollutant – mercury. The primary purpose of this document is to provide information useful to those involved in the control of emissions of mercury from industrial sources. The language and approach are largely technical, but the first two sections should be of interest and value to the general reader.

The requirement to publish this document was established when the Administrator of the Environmental Protection Agency listed mercury as a hazardous air pollutant by notice in the *Federal Register* (Vol. 36, page 5931) on March 21, 1971. The Administrator acted under the authority granted him by Section 112 of the Clean Air Act, which defines a hazardous air pollutant as, "...an air pollutant to which no ambient air quality standard is applicable and which in the judgment of the Administrator may cause, or contribute to, an increase in mortality or an increase in serious irreversible, or incapacitating reversible, illness."

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ABSTRACT

The toxicity of mercury, combined with its high volatility, creates a potential health hazard. This publication deals with two sources of mercury emissions, the primary mercury processing industry and the mercury-cell chlor-alkali industry. An effort is made (1) to identify the process steps that may produce atmospheric mercury emissions, (2) to summarize the emission control techniques and low mercury emission processes used or applicable to these industries, and (3) to evaluate these techniques in terms of cost and effectiveness.

The condenser gas stream is the major source of mercury emissions from a primary mercury processing plant. The amount of emissions can be reduced by converting to processes that inherently produce fewer emissions or by treating effluent gases to remove mercury. Process changes that inherently produce fewer emissions include beneficiation of ore, retort processing, and hydrometallurgical processing. Appropriate control techniques include cooling and mist elimination, wet scrubbing, or adsorption beds.

Major emissions of mercury from a chlor-alkali plant using mercury cells are from the hydrogen gas stream, the end-box ventilation stream, and the cell room ventilation air. The emissions from all sources can be eliminated by converting to the diaphragm-cell process. The cost of converting a 100-ton-per-day plant is estimated to range from $3,700,000 to $8,000,000.

Mercury emissions can also be reduced by the installation of control systems and the use of good housekeeping practices. The hydrogen gas and the end-box ventilation air streams can be treated by cooling and mist elimination, chemical scrubbing, or adsorption beds. No techniques are presently available to treat the cell room ventilation air; therefore, the control of mercury emissions from this source is dependent on good housekeeping practices.

A control system for a primary mercury facility using cooling (down to 45° to 55° F) and mist elimination would cost between $86,000 and $108,000, depending upon the type of mist elimination device used. The cost of a similar control system for a chlor-alkali plant is estimated at $202,000. Chemical scrubbing, which is too expensive for existing primary mercury facilities, can be applied to the chlor-alkali process at a cost from $160,000 to $350,000 for a 100-ton-of-chlorine-per-day plant.

The cost of a carbon bed adsorption system for a primary mercury facility is estimated at $66,000. The capital investment for an adsorption bed system for a chlor-alkali plant of 100-tons-per-day capacity would range from $279,000 to $349,000.

Key words: mercury emissions, chlor-alkali plants, control techniques, costs.

SUMMARY

BACKGROUND INFORMATION

Mercury, one of the most volatile of all metals, vaporizes readily at room temperature. Accordingly, the exposure of a mercury surface to a gas will result in vaporous mercury emissions. This characteristic, combined with mercury's toxicity, may create a potential health hazard in some instances. The saturation concentration of mercury vapor in a gas can be estimated from vapor pressure data and the ideal gas law and used in estimating the vaporous emissions from gas streams containing mercury.

The most important deposits of mercury are found in Italy, Spain, the U.S.S.R., Yugoslavia, China, the United States, Canada, Mexico, and the Philippines. The major deposits of mercury in the United States occur in the West with California and Nevada being the major mercury-producing states. The United States ranked fifth in 1970 and fourth in 1967 through 1969 in the world production of mercury.

MERCURY EMISSIONS, CONTROL TECHNIQUES, AND COSTS

Mercury emissions from process gas streams are dependent on the volumetric flow rate and the concentration of mercury in the gas stream; therefore, appropriate control techniques include cooling the gas, reducing the mercury vapor concentration by chemical scrubbing or adsorption, and reducing the volumetric flow rate of the gas stream.

Mercury emissions can also occur when mercury is exposed to circulating ventilation air. The resultant mercury concentrations are low, but large emissions of mercury can occur where large volumes of ventilation air are used.

Primary Mercury Processing

Mercury can be extracted from ore by two basic methods, pyrometallurgical processing and hydrometallurgical processing. The former process employs heat to volatilize mercury from the ore, whereas the latter method uses an aqueous chemical solution to extract the mercury from the ore.

In the United States, most of the primary mercury is produced through a directly heated pyrometallurgical process. Mercury ore is directly heated by combustion gases in a multiple-hearth or rotary furnace to volatilize mercury. The hot mercury-laden gases leaving the furnace are cooled in an air-cooled heat exchanger, where mercury vapor condenses and collects under an aqueous layer. The condenser gas stream is then emitted to the atmosphere through a stack, which is the major mercury emission point in the process. A typical emission from condenser stack gases is approximately 25 pounds of mercury per day for an existing 100 ton-of-ore-per-day direct-heat extraction process in which the exit gas temperature is 110° F.

In another application of a pyrometallurgical process, mercury ore is heated indirectly by combustion gases to volatilize mercury in a retort. This batch process is generally used in small operations to process high-grade or concentrated ore. The mercury vapor that is evolved is condensed in a water- or air-cooled heat exchanger and collected under an aqueous layer. The major mercury emission from an indirectly heated retort probably occurs while the retort is being charged and discharged. The condenser stack gas flow rate is low as a result of the indirect heating method; therefore, the stack emissions from a retort process are low.

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Mercury can be leached from mercury ore with an aqueous solution of sodium sulfide or sodium sulfite-sodium hydroxide, or by electro-oxidation in an aqueous sodium chloride solution. The soluble mercury compounds that are formed can be precipitated with metals such as zinc, iron, and aluminum, or electro-deposited to recover the mercury. Atmospheric mercury emissions from these hydrometallurgical processes are negligible. A potential water pollution problem exists, however, because soluble mercury compounds may be emitted with the tailings and leach solution. Currently, there are no commercial hydrometallurgical processes in operation in the United States, but the process has been investigated on the research and development level.

The primary mercury industry currently employs little if any control technology to reduce mercury emissions from the condenser stack gases. As a result, few of the techniques that are discussed as control methods have actually been used in this industry. Cooling, mist elimination, water scrubbing, and adsorption are control techniques that have been used successfully in reducing mercury emissions from similar gas streams and should be applicable in reducing emissions from primary mercury extraction plant effluents.

Chlor-Alkali Processing

Chlorine gas and alkali metal hydroxide can be produced in electrolytic cells by the diaphragm-cell process or the mercury-cell process. In the diaphragm-cell process, an asbestos diaphragm separates the anode from the cathode. Chlorine gas is formed at the anode and hydrogen gas and caustic are formed at the cathode by the electrolytic decomposition of a salt solution. Approximately 72 percent of the domestic chlorine is produced by this process. One disadvantage of this process, compared with the mercury-cell process, is that the diaphragm cell produces a low-grade caustic that must be concentrated and purified. This process, however, produces no mercury emissions.

Mercury is used as a flowing cathode in the mercury-cell process. The mercury electrolytic cell is composed of the electrolyzer and the decomposer. In the electrolyzer section, a salt solution, usually NaCl, flows cocurrently with the mercury cathode. A high current density is applied between the mercury cathode and the carbon or metal anodes. Chlorine gas forms at the anode and alkali amalgam forms at the cathode. The amalgam is separated from the brine in a discharge end-box and enters the decomposer section, where water is added. In the decomposer, the amalgam becomes the anode to a short-circuited graphite cathode. Hydrogen gas and alkali metal hydroxide are formed in the decomposer, and the amalgam is converted back to mercury. The mercury is then recycled to the inlet end-box, where it reenters the electrolyzer. The major emissions of mercury from this process occur with the hydrogen gas, the end-box ventilation system, and the cell room ventilation air.

After leaving the decomposer, the hydrogen gas stream contains substantial mercury and water vapor. This stream is usually cooled in one or two stages. The mercury that is condensed from this cooling operation is often removed by a mist elimination device before the hydrogen is vented to the atmosphere or burned as fuel. In some cases, the hydrogen is subjected to additional treatment such as passage through an adsorption bed or scrubbing device.

The end-box ventilation air may consist of air ventilated from the end-boxes, the mercury pump sumps, and the water collection tank. This gas stream may or may not be saturated with mercury vapor, depending on the individual plant. The stream is normally cooled with ambient water before it is emitted to the atmosphere. Additional treatment of the end-box ventilation system consists of methods similar to those applied to the hydrogen stream.

The cell room is ventilated in order to cool the cell room and also to dilute the concentration of mercury to protect the

health of cell room workers. For a plant producing 100 tons of chlorine per day, the ventilation air flow rate for the cell room may vary from 100,000 to 1,000,000 cubic feet per minute. Although a low cell room mercury concentration is usually maintained (50 to 100 micrograms per cubic meter), the high volumetric flow rate can cause the cell room ventilation air to be a major source of mercury emissions from a chlor-alkali plant. No applicable control techniques are currently available for the reduction of mercury emissions from low-concentration, high-volume gas streams. Meticulous housekeeping and maintenance procedures should be employed to minimize the amount of mercury vapor entering the cell room atmosphere.

Emission Control Techniques

Mercury emissions can be reduced by treating effluent gases to remove mercury or by converting to processes that inherently produce fewer emissions. Conversion from direct to indirect heating of ore, beneficiation of ore, and conversion from pyrometallurgical to hydrometallurgical processing are methods that can reduce atmospheric mercury emissions from primary mercury production facilities. The conversion of a mercury-cell process to a diaphragm-cell process will eliminate the mercury emissions from chlor-alkali production plants.

The cooling of a gas stream containing mercury vapor below its mercury saturation temperature will cause mercury vapor to condense, thereby reducing the concentration of mercury vapor in the gas. Some of the condensed mercury remains in the gas stream as a fine mist and can be removed with a mist elimination device or a water scrubber to increase the efficiency of the cooling technique.

Mercury vapor can be removed from a gas stream by adsorption and chemical scrubbing techniques. Molecular sieve and activated-carbon beds can be used to adsorb mercury vapor from a stream. Mercury vapor can also be removed chemically by scrubbing the gas with hypochlorite, depleted brine, or hot concentrated sulfuric acid solutions.

Costs

Costs are presented for those control systems applicable to both primary mercury processing facilities and chlor-alkali plants. The costs of control systems given for primary mercury facilities are for a processing operation having a capacity of 100 tons of ore per day. Control system costs for chlor-alkali plants are based on the control of emissions from both the hydrogen stream and the end-box ventilation stream for a plant having a capacity of 100 tons of chlorine per day.

A control system for a primary mercury facility using cooling to 45° to 55°F and mist elimination would cost between $86,000 and $108,000, depending upon the type of mist elimination device used. The cost of a similar control system for a chlor-alkali plant is estimated to be approximately $202,000.

Chemical scrubbing, preceded by at least partial cooling and mist elimination, is a control system applicable to both source categories. Because of the expense, however, this type of control would not be feasible for existing primary mercury facilities. Costs for a chlor-alkali plant having a capacity of 100 tons of chlorine per day are estimated to range from $160,000 to $350,000.

A third class of control system uses adsorption beds of either treated activated carbon or a molecular sieve/adsorbent blend and is applicable to both primary mercury facilities and chlor-alkali plants. The cost of a carbon bed system for a 100-ton-per-day primary mercury facility is estimated to be $66,000. A molecular sieve system would not be feasible for the control of emissions from this source because of the expense. The capital investment for an adsorption bed system for a chlor-alkali plant of 100-ton-per-day capacity would range from $279,000 to $349,000.

CONTROL TECHNIQUES FOR MERCURY EMISSIONS FROM EXTRACTION AND CHLOR-ALKALI PLANTS

1. INTRODUCTION

The hazardous nature of mercury has been known for centuries. Mercury is hazardous to public health chiefly because of its interference with the central nervous system. In addition, mercury is volatile, so that mercury in the ambient air may approach hazardous levels.

The scope of this document is specifically limited to consideration of mercury emissions and control techniques for those emissions in the primary mercury processing industry and in the mercury-cell chlor-alkali industry.

The objectives of this report are (1) to present, in a concise manner, the essential details of the operation of each industry; (2) to identify the process steps that may produce atmospheric mercury emissions; (3) to summarize the emission control techniques and low-mercury-emission processes used in or applicable to these industries; and (4) to evaluate these techniques in terms of cost and effectiveness.

The information used in the preparation of this document was obtained from four sources. An extensive review of the literature was conducted with emphasis placed on recent publications and reports. This material was evaluated and considered together with comments from management and trade-association personnel and information developed by contractors. Plant visits provided insight into current emission control practices and applicable control techniques and processes. Finally, a limited amount of source testing was performed to determine plant emission levels and to evaluate the effectiveness of control techniques.

The information contained in this document is intended to give an appraisal of the emission control methods, techniques, and processes currently being used in or potentially adaptable to primary mercury extraction plants and mercury-cell chlor-alkali plants.

2. BACKGROUND INFORMATION

2.1 DEFINITIONS

2.1.1 Primary Mercury Extraction

Beneficiation – Methods by which low-grade ore is processed into higher-grade ore or concentrate.

Cinnabar – Mercury-bearing (in the form of mercuric sulfide, HgS) mineral that is the primary source of the metal.

Condenser stack gas – Vapor and particulate matter emitted from the condenser stack after having passed through a condenser system.

Directly heated furnace – Furnace in which ore is heated directly by hot combustion gases.

Hoeing operation – Process whereby mercury, dust, and soot collected in the condenser system of a primary plant are mixed with lime and alternately scraped and pressed by mechanical or manual means to cause coalescing of minute mercury droplets.

Hydrometallurgical process – Procedure that uses a water solution of various chemicals to extract metal from its ore.

Indirectly heated furnace – Furnace in which ore is heated indirectly by combustion gases; that is, the combustion gases are never in contact with the ore.

Particulate matter – Any material, except uncombined water, that exists as a solid or liquid in the atmosphere or in a gas stream.

Primary mercury production – Production of mercury metal from mercury-bearing ore.

Pyrometallurgical process – Procedure that uses heat to extract metal from its ore.

Retort – Type of indirectly heated furnace.

2.1.2 Chlor-Alkali Plants

Amalgam – Alloy of mercury and another metal, such as sodium or potassium.

Anode – Positive pole of an electrolytic cell.

Cathode – Negative pole of an electrolytic cell.

Contact cooler – Tower in which a liquid is used for direct contact cooling of a gas stream.

Denuder or decomposer – Device into which the sodium or potassium amalgam from the electrolyzer flows continuously. Within this device, deionized water or other chemicals are added, and the amalgam becomes the anode to a short-circuited iron or graphite cathode in an electrolyte of sodium or potassium hydroxide solution. Hydrogen gas is formed and the sodium or potassium hydroxide is increased to a 50 percent solution. Regeneration of the mercury occurs simultaneously, and the mercury is recycled back to the chlorine cell for continuous usage.

Diaphragm cell – Electrolytic device employing porous asbestos coating over the cathode screen that separates the chlorine gas evolved at the anode from the hydrogen gas evolved at the cathode.

Mercury-cell chlor-alkali electrolyzer – Electrolytic device that uses a flowing mercury cathode to make chlorine gas and sodium or potassium amalgam.

Mercury knockout drum – Device with a tangential top inlet for process gases containing mercury vapor, mercury mist, and water. As the gas spirals downward, centrifugal forces separate some of the mercury mist and water droplets from the process gas stream. Mercury collects at the bottom under a layer of water while cleaned gas flows out of a pipe in the top of the drum.

Mercury mist eliminator – Device that uses direct impingement or high centrifugal forces for the removal of mercury mist from process gas streams.

2.2 PROPERTIES OF MERCURY

Mercury is found in nature in many forms, but the principal mercury ore is cinnabar. Cinnabar in the pure form is red, has a specific gravity of 8.1, and contains 86.2 percent mercury by weight. In the United States, the average mined ore contains approximately 5 pounds of mercury per ton of ore.

Mercury is the only common metal that exists as a liquid at ordinary temperatures.[1] It is a heavy (molecular weight = 200.59), silver-white, shining metal at normal temperatures but tarnishes at elevated temperatures near its boiling point because of the formation of its oxide, HgO. Mercury combines with many metals to form alloys called amalgams. Mercury also is a fair conductor of electricity and has a regular coefficient of expansion. The latter properties make it generally useful in thermometers, barometers, and other instruments.

Some of the properties of elemental mercury are summarized in Tables 2-1 and 2-2. Mercury is a unique metal because of its high vapor pressure. This characteristic, in combination with its toxicity, may create a potential health hazard in some instances. The vapor pressure of mercury for several temperatures is given in Table 2-3.

Table 2-1. ISOTOPIC ABUNDANCE OF MERCURY[1]

Isotope, mol wt	Abundance, percent
204	6.8
202	29.8
201	13.2
200	23.1
199	16.8
198	10.0
196	0.15

Table 2-2. PHYSICAL PROPERTIES OF ELEMENTAL MERCURY[1]

Property	Value and units
Avg mol wt	200
Density	13.595[5] g/ml at 0°C
Surface tension	484 dynes/cm vac 20°C
Heat of fusion	2.7 cal/g
Heat of vaporization	13.985 kcal/g mole
Melting point	–38.9°C
Boiling point	356.6°C

Table 2-3. VAPOR PRESSURE OF MERCURY[1]

Temperature, °C	Vapor pressure, mm Hg
–10	0.0000606
0	0.000185
20	0.001201
40	0.006079
60	0.02524
80	0.08880
100	0.2729

If mercury vapor is assumed to be an ideal gas, the equilibrium concentration of mercury vapor in a gas can be calculated using vapor pressure data. Table A-1 and Figure A-1 in the Appendix give this information.

2.3 ORIGIN, PRODUCTION, AND USES OF MERCURY

2.3.1 World

Although there are 25 known mercury-bearing minerals, the primary source of this metal is cinnabar (mercuric sulfide).[2] Other economically important mineralogical species of mercury include the sulfide minerals of iron, arsenic, and antimony.

The gangue associated with cinnabar deposits includes carbonate and silicate minerals such as calcite, chalcedony, dolomite, opalite, quartz, and serpentine.[2] The deposits usually are shallow, extending downward to depths of slightly less than 2500 feet. Almost

all of the deposits are in areas of tertiary or quaternary volcanic activity. The most important deposits occur in Italy, Spain, the U.S.S.R., Yugoslavia, China, the U.S., Canada, Mexico, and the Philippines.

The annual world production of mercury by country and the average London-based price per flask from 1968 through 1970 are given in Table 2-4.

Table 2-4. WORLD PRODUCTION OF MERCURY BY COUNTRY FOR 1968 THROUGH 1970[3] (76-pound flasks[a])

Country	Year		
	1968	1969	1970
Bolivia (exports)	134	68	12
Canada	5,700	21,200	24,400
Chile	513	286	380[b]
China, mainland[b]	20,000	20,000	20,000
Colombia	362	344	350[b]
Czechoslovakia	116	435	2,000[b]
Ireland	.....	420	1,604[c]
Italy	53,317	48,733	44,382
Japan	5,084	6,543	5,907
Mexico[d]	17,202	22,539	30,269
Peru	3,132	3,365	3,400[b]
Philippines	3,544	3,478	4,648
Spain	56,943	64,862	47,689
Tunisia	309	244	100
Turkey	4,670	6,566	8,592
U.S.S.R.[b]	45,000	47,000	48,000
United States	28,874	29,640	27,303
Yugoslavia	14,794	14,330	15,461
Total[e]	259,694	290,043	284,497

[a]Average price per 76-lb flask, London = $546.80 in 1968; $536.41 in 1969; and $411.45 in 1970.
[b]Estimate.
[c]Sales only.
[d]Official figures as reported by Statistical Office, Secretary of Industry and Commerce, Mexico; overall production of mercury believed to be much higher.
[e]Total is of listed figures only.

2.3.2 United States

The major mercury deposits in North America are located in the western part of the continent (see Figure 2-1). A summary of the United States mercury statistics is given in Table 2-5. These statistics indicate a considerable decrease in U.S. mercury production in 1971 over previous production, which probably results from decreased demand and lower prices. Production decreased from 27,303 flasks in 1970 to an estimated 17,445 flasks in 1971. During this same period, prices fell from a 1970 average of $408 per flask to a 1971 year-end price of $218 per flask.

California mines accounted for more than 85 percent of the domestic production of mercury during the first 50 years of the twentieth century.[3] During recent years, California and Nevada have been the major producing states. The production in the United States by states for 1969 and 1970 is summarized in Table 2-6.

Mercury consumption in the United States generally increased from 1950 to 1969 but declined significantly in 1970 and 1971. Table 2-7 summarizes the consumption of mercury in the United States, by use, from 1950 to 1971. The major mercury uses in recent years have been in electrical apparatus, electrolytic preparation of chlorine and caustic soda, and mildew-proofing for paints. The future use of mercury in the electrolytic preparation of chlorine and caustic soda and as a mildew-proofing for paints is expected to be substantially reduced by recently proposed Environmental Protection Agency control actions. Figure 2-2 gives trends in production, consumption, and price of mercury.

2.4 EMISSION MECHANISMS AND GENERALIZED CONTROL PRINCIPLES

Elemental mercury has unusually high volatility for a metal. This property can lead to high emissions of mercury in process gas streams and in room ventilation air.

2.4.1 Process Gases

Gases present in various processes involving mercury tend to become saturated with mercury vapor. These gases, if vented without treatment, carry the mercury vapor into the atmosphere. The amount of mercury emitted in this manner is independent of the kind of gas involved, but depends upon: (1) the temperature of the system, (2) the degree of saturation of the gas, and (3) the volumetric flow rate of the gas.

These dependency factors suggest that appropriate control procedures consist of: (1) cooling the gas to condense mercury, (2) reducing the mercury vapor concentration

Figure 2-1. Mercury deposits in North America.[2]

Table 2-5. UNITED STATES MERCURY STATISTICS[3]

	Year					
	1966	1967	1968	1969	1970	1971[a]
Producing mines	130	122	87	109	79	30
Production, 76-lb flasks	22,008	23,784	28,874	29,640	27,303	17,445
Price,[b] $	441.72	489.36	535.56	505.04	407.77	292.41[c]
Value, $10[3]	9,722	11,639	15,464	14,969	11,134	------
Exports, flasks	357	2,627	7,496	507	4,653	7,232
Re-exports	476	475	103	108	50	------
Imports:						
For consumption	31,364	24,348	23,246	31,924	21,972	------
General	34,757	23,899	23,956	30,848	21,672	29,732
Stocks Dec. 31	20,076	18,277	22,907	22,692	16,376	------
Consumption	71,509	69,517	75,422	77,372	61,503	52,725

[a]1971 preliminary estimates.
[b]Average price per flask, New York.
[c]Year-end price per flask = $218.

Table 2-6. MERCURY PRODUCED IN THE UNITED STATES, BY STATE FOR 1969 AND 1970[3]

State	Producing mines		Flasks[a]		Value,[b] 10^3	
	1969	1970	1969	1970	1969	1970
California	72	51	18,480	18,593	9,333	7,583
Idaho	1	1	1,012	1,038	511	423
Nevada	24	13	8,165	4,916	4,124	2,005
Oregon	4	5	43	274	22	112
Alaska, Arizona, New York, and Texas	8		1,940		979	
Alaska, Arkansas, New York, Texas, and Washington		9		2,482		1,012
Total	109	79	29,640	27,303	14,969	11,134

[a]For 76-pound flasks.
[b]Value calculated at average New York price.

through chemical scrubbing or adsorption, and (3) reducing the volumetric flow rate.

In order to estimate the quantity of mercury emitted, equilibrium (that is, complete saturation) is usually assumed. The results represent the maximum emission of mercury if the gas stream is not saturated, but more frequently a minimum emission if the gas stream is saturated and also contains mercury particulates. The required calculations are presented in detail in the Appendix together with a table of calculated results.

2.4.2 Ventilation Air

Mercury emissions can occur in ventilation air if mercury is exposed to the circulating air either as a result of uncovered containers or through incomplete clean-up of spills. In this case, the concentration of mercury vapor is much lower than the saturation value; but because of the large volumes of ventilation air used, relatively large emissions of mercury can occur. The amount of mercury emitted, E, is the product of the ventilation air volumetric flow rate and the average mercury vapor concentration. The following expression may be used:

$$E = 0.09 \times 10^{-6} \times V \times C$$

where: E = mercury emission, pounds per day
V = ventilation volumetric flow rate, cubic feet per minute
C = mercury vapor concentration, micrograms per cubic meter.

The dependency factors cited in Section 2.4.1 indicate the appropriate approaches to the control of mercury emissions in ventilation air; namely, to reduce the volume of the air or to reduce the concentration of mercury vapor in the air. Because the ventilation air volume is usually dictated by area cooling requirements, reduction of concentration remains the only recourse. The concentration can be reduced by minimizing the sources of exposed mercury through careful handling and good housekeeping.

Frequently, the concentration of mercury vapor in the ventilation air is not known. A conservative estimate of this mercury emission may be made by assuming the vapor concentration to be equal to the current Threshold Limit Value (TLV) set by the American Conference of Governmental Industrial Hygienists (ACGIH), which is 50 micro-

Table 2-7. MERCURY CONSUMED IN THE UNITED STATES[3,4,5]
(76-pound flasks)

Use	Year								
	1950	1960	1965	1966	1967	1968	1969	1970	1971
Agriculture[a]	4,504	2,974	3,116	2,374	3,732	3,430	2,689	1,811	1,478
Amalgamation	192	255	268	268	219	267	195	219	-----
Catalysts	2,743	1,018	924	1,932	2,689	1,914	2,958	2,238	996
Dental preparations	1,458	1,783	1,619	1,334	1,359	2,089	2,880	2,286	1,871
Electrical apparatus	12,049	9,268	16,097	16,257	14,610	17,484	18,490	15,952	16,646
Electrical preparation of chlorine and caustic soda	1,309	6,211	8,753	11,541	14,306	17.458	20,720	15,011	12,252
General laboratory	646	1,302	1,119	1,563	1,133	1,246	1,936	1,806	1,357
Industrial and control instruments	5,385	6,525	4,628	4,097	3,865	3,935	6,655	4,832	3,906
Paint									
Antifouling	3,133	1,360	255	140	152	392	244	198	414
Mildew-proofing	-----	2,861	8,211	8,280	7,026	10,174	9,486	10,149	8,192
Paper and pulp manufacturing	-----	3,481	619	612	446	417	588	226	2
Pharmaceuticals	5,996	1,729	418	232	283	424	712	690	668
Redistilled[b]	7,600	9,678	-----	-----	-----	-----	-----	-----	-----
Other[c]	4,200	2,722	15,402	15,632	12,568	7,945	9,134	5,858	2,292
Total identified uses	-----	-----	73,560	71,509	69,517	75,422	76,657	61,276	50,074
Total unidentified uses	-----	-----	-----	-----	-----	-----	715	227	-----
Grand total[d]	49,215	51,167	73,660	71,509	69,517	75,422	77,372	61,503	52,725

[a]Includes fungicides and bactericides for industrial purposes and, prior to 1959, also includes pulp and paper manufacturing.
[b]"Redistilled" used in industrial instruments, dental preparations, and electrical apparatus.
[c]"Other" includes mercury used for installation of chlor-alkali plants for 1963 and later dates.
[d]All items do not add up to the total given, which has been increased to cover approximate total consumption.

grams per cubic meter. For example, assuming that the concentration does not exceed the current TLV and using a ventilation flow rate of 1 million cubic feet per minute of air, the estimated cell room emission rate would be 4.5 pounds per day.

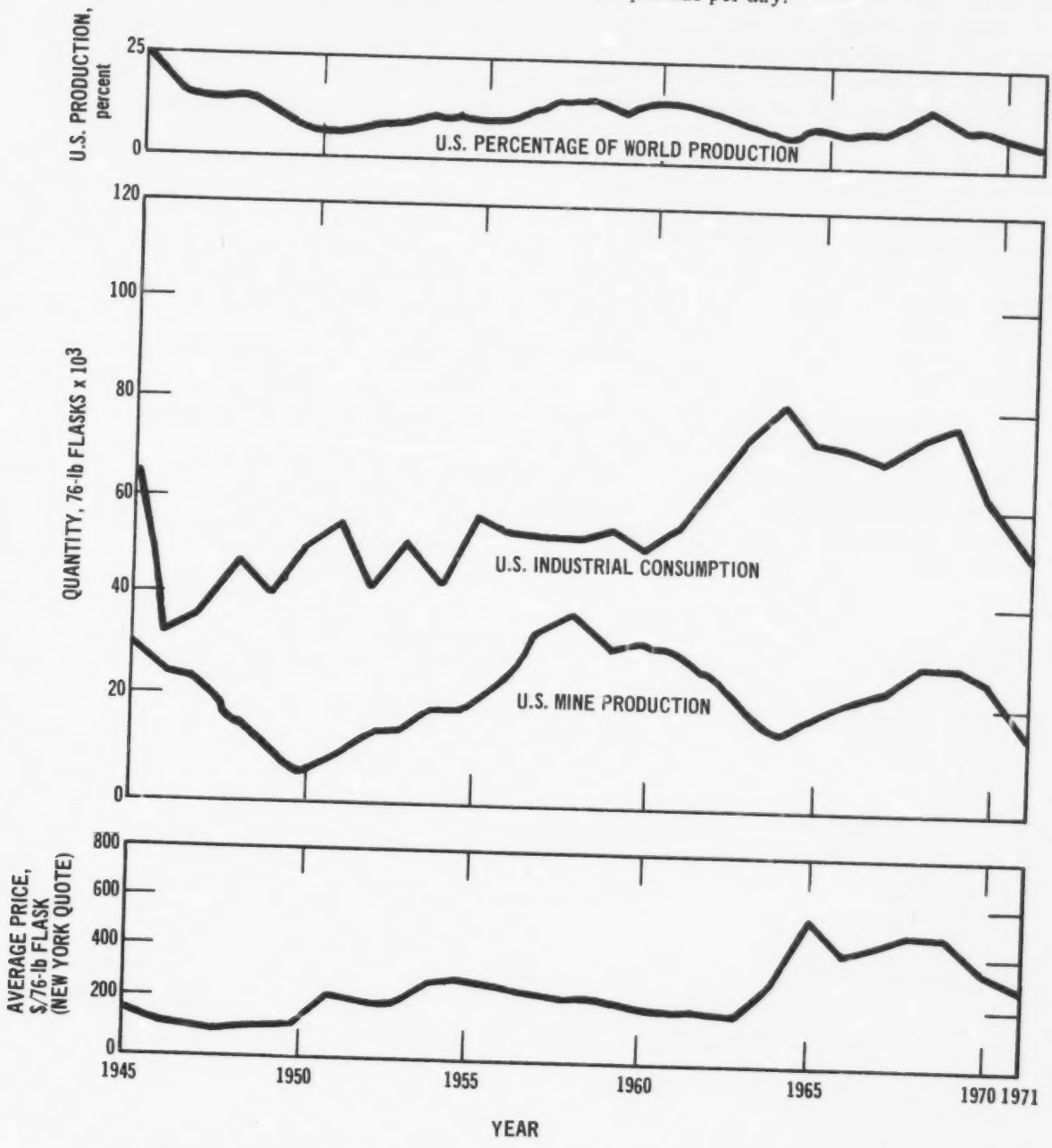

Figure 2-2. Trends in U. S. production, consumption, and price of mercury.[3] Values for 1971 are preliminary.

2.4.3 Particulate Emissions

In addition to vapor losses, mercury emissions can occur in the particulate form as elemental mercury mist, solid mercury compounds, and mercury adsorbed on soot. Particulate emissions are frequently encountered when a hot gas saturated with mercury vapor is cooled. The mercury tends to condense in the form of a mist that may be entrained in the gas stream and carried to the atmosphere, thus obviating the effect of the gas cooling step.[2] Amounts of particulate mercury emissions cannot be readily predicted. Control consists of collection and retention of the particles involved by means of an entrainment separator, often called a mist eliminator.

It will be seen in the following discussions of each specific industry that there is a commonality of principle inherent in control practice but not necessarily a commonality of application because of the system variables encountered.

2.5 REFERENCES FOR SECTION 2

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3. Cammarota, V.A., Jr. 1970 Bureau of Mines Minerals Yearbook, Mercury (Preprint). U.S. Department of Interior, Bureau of Mines. Washington, D.C. 1971. p. 1, 3-5, 8.
4. U.S. Bureau of Mines Minerals Yearbook, Mercury. U.S. Department of Interior, Bureau of Mines. Washington, D. C. 1950-1968.
5. Mercury in the Fourth Quarter of 1971. U.S. Department of Interior, Bureau of Mines. Washington, D.C. February 22, 1972. 5 p.

3. MERCURY EMISSIONS, CONTROL TECHNIQUES, AND CONTROL COSTS

3.1 PRIMARY MERCURY PRODUCTION

3.1.1 Emissions and Process Description

3.1.1.1 Rotary Furnaces

3.1.1.1.1 Process Description – Most of the mercury produced in the United States is extracted by the use of directly fired rotary furnaces. A diagram of a typical rotary furnace operation is shown in Figure 3-1.[1] This type of furnace generally ranges from 2 to 7 feet in diameter and 24 to 140 feet in length; the ore treatment capacity ranges from 10 to 300 tons of ore per day.[2-6]

Crushed mercury ore ranging in size from 3/4 to 4 inches in diameter is fed into the rotary furnace by means of a reciprocating tube-type feeder known as a "shotgun" feeder. The ore feed rate is adjusted with this device to obtain ore temperatures of 1100° to 1600°F within the furnace.[2-6] The proper furnace retention time for a particular ore may vary from 15 to 60 minutes. The ore retention time is determined by the inclination slope of the furnace, which varies from 1/2 to 3/4 inch per foot, and by the rate at which the furnace rotates, generally between 1/2 and 3-1/2 revolutions per minute.[2-6] The calcined ore is discharged from the furnace into calcine or burnt ore bins, from which it is subsequently transported to a waste dump. The hot calcined ore is used in some cases to preheat the combustion air or the feed ore.

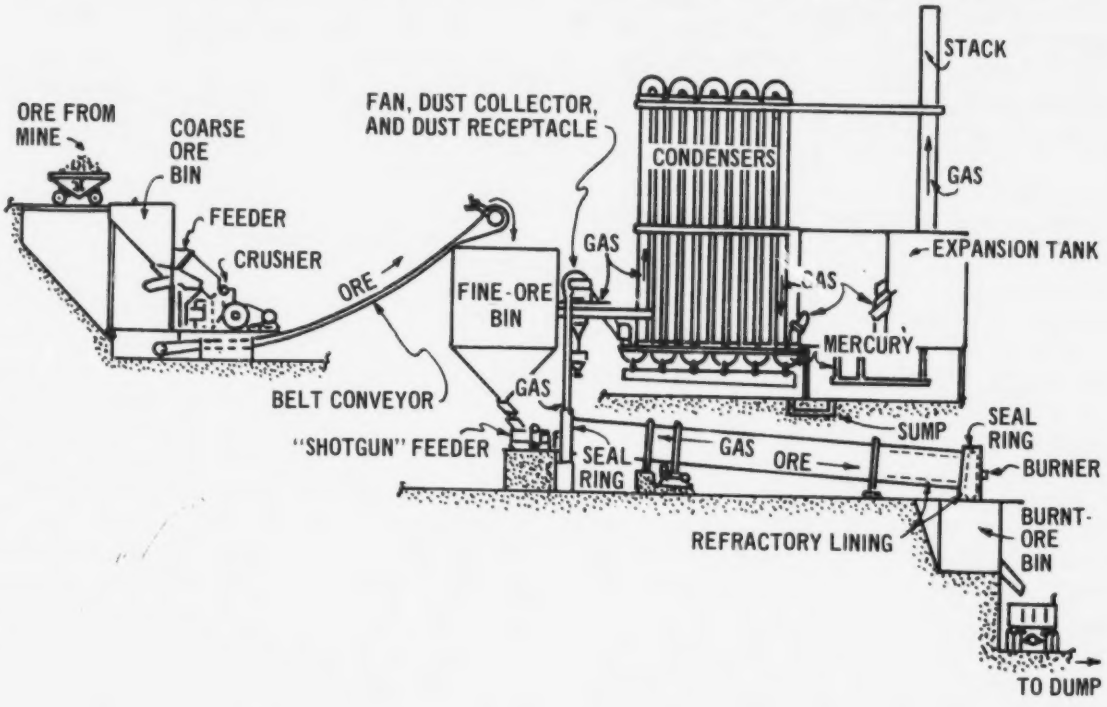

Figure 3-1. Pyrometallurgical process for producing mercury.[1]

Oil is usually burned as fuel with approximately 50 percent excess air; however, if the oil is sufficiently preheated and atomized, 35 percent excess air can be used.[6,7] The rate of oil use generally ranges from 7 to 10 gallons of oil burned per ton of ore furnaced. If natural gas is used, the excess air used can be reduced to 25 percent because of the more efficient combustion of this fuel.[7]

The hot combustion gases flow countercurrently to the ore and heat the ore to temperatures that volatilize the mercury. The mercury-bearing vapors leave the furnace at temperatures ranging from 450° to 600°F and pass through one or more cyclone separators, which remove most of the particulate matter. The dust collectors are operated at 450° to 500°F to avoid condensation of the mercury vapor.[2-6,8] A blower on the downstream side of the cyclone maintains a slight suction on the furnace to minimize leakage of hot mercury-laden gases from the furnace.

After passage through the cyclone and blower, the gas stream is usually divided and introduced into sufficient banks of air-cooled verticle U-tube condensers to control its flow rate and temperature. The condenser pipes are about 16 inches in diameter and from 20 to 40 feet in height.[4-6,8] The condensers are constructed of cast iron, mild steel, tile, stainless steel, Monel,* or fiber glass, depending on the sulfur or chloride content of the ore and the subsequent corrosiveness of the gases. The individual pipes are connected at the top and bottom to adjacent pipes. The lower ends of consecutive condenser pipes are connected and sealed with water in a container called a launderer, or in the case of larger capacity plants, a condenser tank. Condensed mercury, dust, and soot fall down the condenser pipes into the launderer, or into buckets that are submerged in the launderer and placed under each connection. Periodically, the condenser pipes are washed down with a water spray to remove the mercury, dust, and soot adhering to the insides of the pipes.

The mercury-bearing mud accumulated in the launderer or in the buckets is periodically collected, transferred to a hoeing table, mixed with lime, and hoed manually or mechanically to collect the mercury. During the hoeing operation, the mud is scraped and pressed, causing minute mercury droplets to coalesce and form larger drops of mercury that flow into a collection tank. The mud remaining after the hoeing operation is completed is processed in a retort furnace or is recycled back into the main ore furnace.

The total length of a condenser system depends on the volume of gas and the amount of cooling desired. The condenser system normally is designed to provide a temperature of less than 110°F out of the stack.[4,5,8] Although the condenser normally depends on natural air convection for cooling, a common practice is to spray water on the outside of the hottest pipes in the condensing system, particularly during the summer months.

After the gases leave the condenser system, they are expanded into one or more wooden tanks designed to maintain the proper system draft.[6] These wooden expansion tanks also provide some additional cooling and mist elimination. From these tanks, the condenser gases pass into the stack from which they are emitted to the atmosphere at temperatures of 90° to 110°F.[2-6,8] A cooling practice that is sometimes employed involves spraying water into the wooden expansion tanks to cool the condenser gases further.

The calculated combustion gas volumetric flow rates for a typical directly heated furnace employing propane or fuel oil with 0, 25, and 50 percent excess air are given in Table 3-1.

As can be seen from Table 3-1, the calculated volume of gases generated from combustion and excess air for the two fuels listed in the table are essentially equal. The use of propane or natural gas as fuel would offer a slight advantage because less soot would be generated and a smaller amount of

* Mention of commercial products or commercial names does not constitute endorsement by the Environmental Protection Agency.

Table 3-1. CALCULATED VOLUMETRIC FLOW RATES (scfm)[a] DUE TO COMBUSTION AND EXCESS AIR FOR A 100-TON/DAY ORE RATE

Fuel	Excess air, %		
Fuel	0	25	50
Fuel oil	800	1000	1200
Propane	900	1100	1300

[a] Standard conditions are 70°F and 29.92 in. Hg.

excess air would be necessary as a result of more efficient combustion. The direct advantages are reduced stack gas emissions and particulate matter formation, which in turn reduce both mercury vapor losses and mercury losses due to adsorption of the metal on soot.

Excess air should be kept to a minimum as dictated by complete fuel combustion and sulfur oxidation inasmuch as the mercury emission rate is dependent on the stack gas volumetric flow rate. Table 3-1 indicates that in going from 50 percent to 25 percent excess air furnace operation, a volumetric flow rate reduction of 15 percent would result.

"Combustion gas," as used in Table 3-1, is the gas, including excess air, that enters the furnace immediately after combustion has occurred. In actuality, other gases, such as water vapor or sulfur dioxide, are driven off the ore or are formed during combustion. These additional gases increase the volume of gas that flows out of the stack. Furnace design calculations for directly fired furnaces generally indicate a total stack volumetric flow rate of 1500 to 1700 standard cubic feet per minute (scfm) per 100 tons of ore per day, assuming that an averaged silica carbonate type mercury ore is being calcined.[6]

3.1.1.1.2 Emissions – The major emissions of mercury from a primary mercury furnacing operation occur from the condenser stack. Other minor emission points are dust and vapor emissions from the mining operation, the furnace room ventilation air, the hoe table ventilation air, and emissions from the hot discharged ore. These minor emissions can be minimized by good housekeeping and operating practices.

The mercury emissions from the condenser stack effluent can vary with the grade and type of ore processed, and with variations in plant operating practices. A typical condenser stack gas is described below:

1. Stack gas temperatures of 90° to 110°F.
2. Stack gas saturated with mercury vapor.
3. Particulate mercury emission equal to the mercury vapor emission. (This is based on source testing results obtained by the Environmental Protection Agency at several mercury extraction facilities.)[9-11]
4. Stack gas volumetric flow rates of 1000 to 1600 standard cubic feet per minute.

Stack emissions are calculated and presented in Table 3-2.

Table 3-2. CALCULATED MERCURY EMISSIONS FROM THE CONDENSER STACK OF A PRIMARY MERCURY EXTRACTION FACILITY

Temperature, °F	Emissions, lb/day	
	At 1000 scfm	At 1600 scfm
90	6.6	10.5
110	15.4	24.7

A condenser stack gas flow rate of 1600 standard cubic feet per minute corresponds to a 100-ton-per-day ore treatment rate; therefore, the estimated emission from a 100-ton-per-day facility with a 110°F stack effluent temperature is approximately 25 pounds per day.

In actual emission sampling tests conducted by the Environmental Protection Agency at three primary mercury extraction facilities, mercury emissions per 100-ton-per-day ore treatment rate ranged between 18 and 59 pounds per day.[9-11]

3.1.1.2 Multiple-Hearth Furnaces

3.1.1.2.1 Process Description – Multiple-hearth furnaces are not in common use in the

United States for primary mercury production; however, in Europe roughly 50 percent of the primary mercury operations employ these furnaces. A large Canadian mining operation is presently using a multiple-hearth furnace to process a flotation concentrate.

The ore treatment prior to furnacing is more elaborate for a multiple-hearth furnace than for a rotary furnace. The ore feed to the furnace can be concentrated by flotation or other beneficiation processes, or it can be furnaced directly as mined. Beneficiation of ore is discussed in Section 3.1.1.3.

A multiple-hearth furnace consists of a series of circular refractory hearths, placed one on top of the other, that are usually enclosed in a steel shell (Figure 3-2). A center vertical shaft rotates arms that mix the ore by moving it in a spiral path across each hearth. The ore is fed to the top hearth, where it moves across to drop-holes before falling to the next hearth. The ore continues its travel from hearth to hearth in this manner until it is discharged from the bottom of the furnace.

Heat can be supplied alternatively by combustion of the charge elements, combustion of fuel in burners on certain hearths (direct firing), combustion of fuel in a separate combustion chamber (indirect firing), and combustion of fuel in muffles (indirect heating); or by heating with electrical resistor elements (indirect heating). In the most common application, hot combustion gases flow countercurrently to heat the ore to temperatures that will volatilize mercury. The ore is heated continually from the top hearth to the bottom hearth. The temperature and atmosphere of each hearth in a multiple-hearth furnace may be closely regulated.[7]

The calcined ore is discharged from the bottom of the furnace at a temperature of approximately 1200°F. The mercury-vapor-laden combustion gas is discharged from the top of the furnace at temperatures ranging from 500° to 600°F. The gas then enters a cyclone where large particulate matter is removed. A fan on the downstream side of the cyclone maintains a slight suction on the furnace, minimizing vapor emissions from the furnace. The condenser system for a multiple-hearth furnace is essentially the same as that used for a rotary furnace.

3.1.1.2.2 Emissions – The condenser stack gas flow rate and temperature are approximately the same for a multiple-hearth furnace as for a rotary furnace; therefore, the stack emissions of mercury are approximately equal. Minor mercury emission points from a multiple-hearth furnace operation are similar to those previously described for a rotary-furnace operation.

If an indirect method were used to heat the multiple-hearth furnace, the volume of gases produced would be much less, consisting primarily of water vapor, a small amount of air, and sulfur dioxide produced by the oxidation of sulfur. This small volume of gas would greatly reduce the condenser requirement, resulting in a large emission reduction. Such furnaces are commercially available but have not been used in the primary mercury industry because of cost.

3.1.1.3 Beneficiation

3.1.1.3.1 Process Description – Conventionally, mined ore is subjected to pyrometallurgical treatment without preliminary concentration; however, on the basis of differences in the physical properties of the mineral species, for example, density and surface characteristics, methods for preliminary concentration have been used in specific instances. These include hand sorting, jigging, tabling, and flotation.[8] Flotation, when feasible, is the most effective of these techniques. Mercury recoveries of 80 to 90 percent can be attained with this method, depending mainly on the type of rock in which the cinnabar is contained.[8,12,13] A typical mercury ore flotation flow sheet is shown in Figure 3-3.[14]

In a flotation process, the ore is normally subjected to a two-stage crushing operation followed by grinding in a ball mill to approximately 65 mesh.[13] Finer grinding (100 mesh

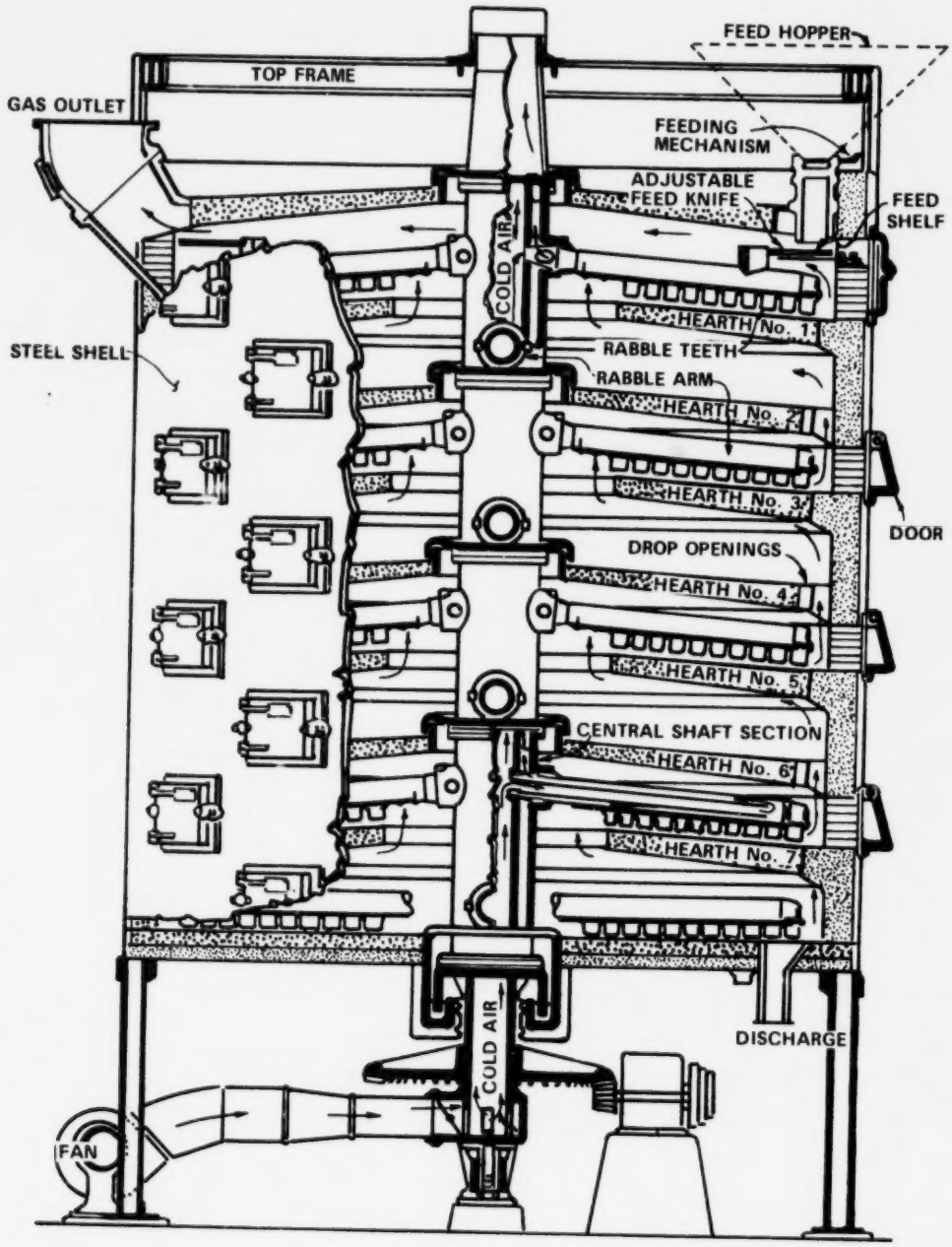

Figure 3-2. Multiple-hearth furnace.[7]

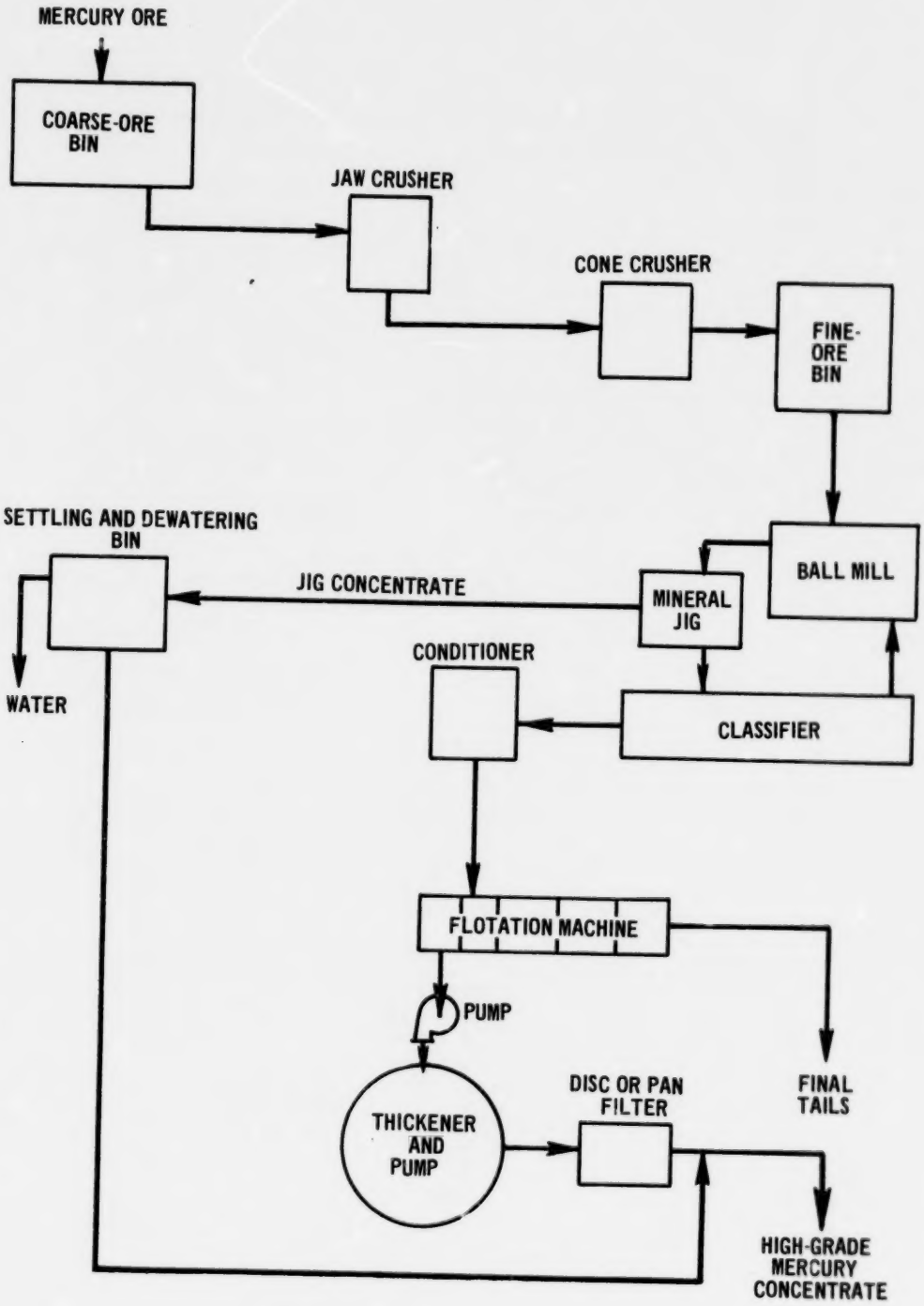

Figure 3-3. Flotation flow sheet for cinnabar ore.[14]

or smaller) may be necessary to improve yields in some ore types. The finely ground ore is then passed through a jigging operation in which the heavier mercury minerals are separated by gravity and are removed as concentrate. The lighter overflow material from the jig is fed to a series of flotation cells where air bubbles float the cinnabar by use of flotation reagents into a concentrate. Concentrates containing from 200 to as high as 1000 pounds of mercury per ton of concentrate are produced. Concentrate is more amenable to retorting, multiple-hearth roasting, and hydrometallurgical processing than to a directly fired rotary-furnace operation.

Flotation and other beneficiation processes are not generally used in the United States in the primary mercury industry. As a general rule, the installed cost for a complete flotation concentration system, exclusive of electrical power and water provisions, is about $3500 per ton of ore processed per day. The operating cost is approximately $5.00 per ton of ore processed.[12,13] The low recovery efficiency and the high initial investment and operating cost have made this method unattractive in the United States.

The water required for flotation ranges from 1.5 to 4.0 tons per ton of ore processed.[12,15] About 70 to 80 percent of this water can be reclaimed if settling ponds are used.[12,15]

Some advantages of flotation are listed below:[14]

1. Furnacing plant operations that employ flotation can be more flexible because of smaller furnace tonnages.
2. Mercury emissions are reduced considerably in directly heated furnace operations since a lower ore treatment tonnage results in a lower furnace stack gas volume per flask of mercury collected. The emission reduction factor is estimated to be roughly equal to the concentration ratio.

3.1.1.3.2 Emissions – The flotation operation, unless properly designed, is a potential source of water pollution. The air emissions from flotation concentration operations are small and may occur as dust emissions during the crushing and grinding operations.

3.1.1.4 Retort

3.1.1.4.1 Process Description – A retort is an indirectly heated furnace and is small in comparison to directly heated furnaces. Retorts are generally classified as pipe retorts or D-retorts, depending on their size and shape.[16]

The pipe retort consists of circular iron pipes 8 to 12 inches in diameter and 7 to 9 feet in length. Several pipes may be collected in banks with one fire box containing up to 12 pipes (Figure 3-4). These pipes can be horizontally situated, in which case the condenser gases are taken from the rear of the retort (Figure 3-4); or they can be inclined, with the mercury vapor evolving from the high side of the pipes (Figure 3-5). The capacity of each pipe is on the order of 5 cubic feet of ore per charge. The furnacing time varies from 12 to 24 hours per charge of ore.

The D-retort, illustrated in Figure 3-6, has a larger capacity than the pipe retort and has a cross section resembling the letter D. The ore charge for a D-retort varies from 5 to 10 cubic feet of ore. The furnacing times are similar to those of the pipe retort described above. Some D-retorts are operated under a slight suction produced by a blower in the retort stack. In another design, a small air flow (10 cubic feet per minute) is blown through the retort to move the mercury vapor to the condenser system.

In operation, a retort is charged with directly mined ore or concentrated ore, after which the retort door is sealed with special clay that prevents mercury vapor leaks. Lime is mixed with the ore to aid in the oxidation of sulfur to sulfur dioxide.[16] The ore charge is heated from 12 to 24 hours at temperatures

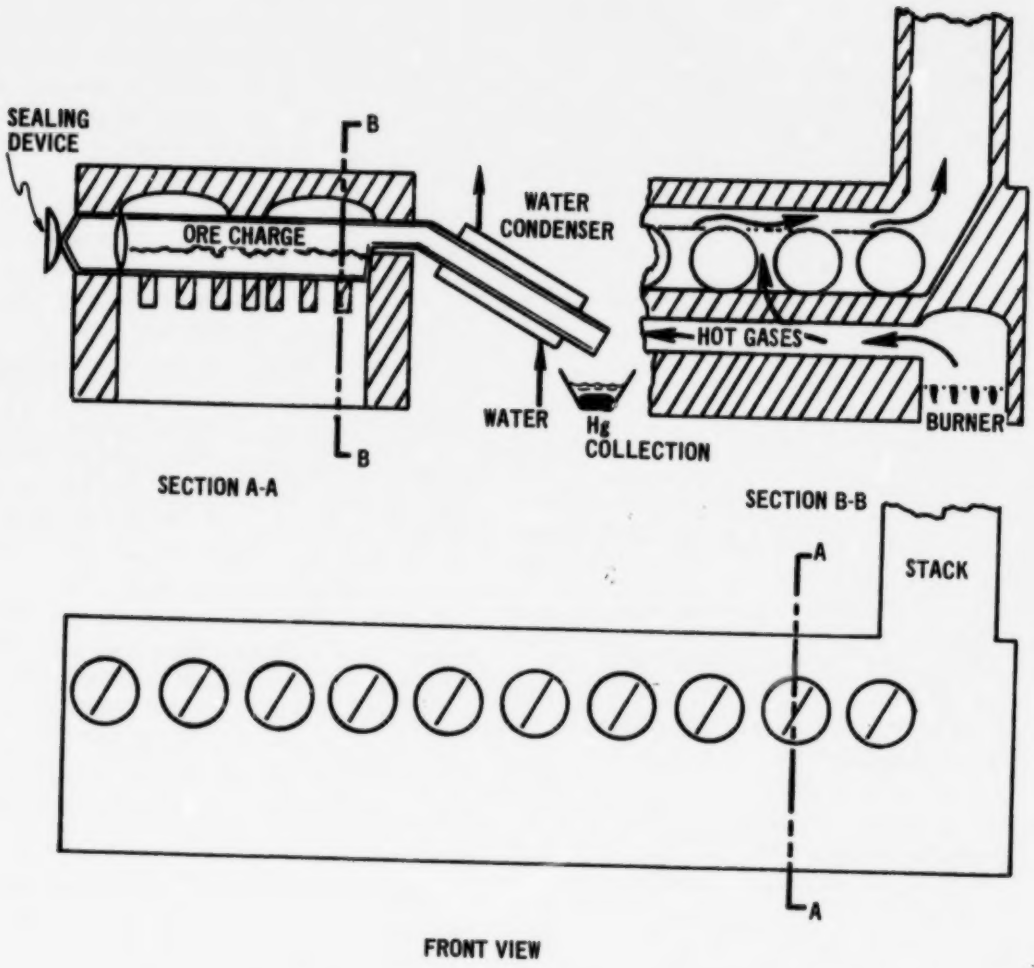

Figure 3-4. Group horizontal pipe retort.[16]

of 1200° to 1400°F. At the end of this heating period, the calcine is removed, and another charge of ore is added.

The mercury-vapor-laden gas enters the condenser system from the retort. The condenser system may be of an air-cooled or an indirect or direct water-cooled type. If water is available, an indirect water-cooled condenser is usually used. A typical water condenser consists of a jacketed 12-foot section of 3- or 4-inch-diameter pipe. The condensed mercury is usually collected under water and in general does not require further purification. In some cases mercury-bearing solids collected in the condenser system are periodically mixed with lime and are hoed to collect the mercury. Retort processing is not readily adaptable to large mining operations because of high labor requirements and low treatment rates.

The stack gas volumetric flow rate from a retort is very small because of the indirect-heating method employed.[11]

3.1.1.4.2 Emissions – Stack emissions from a retort are small because the stack gas flow

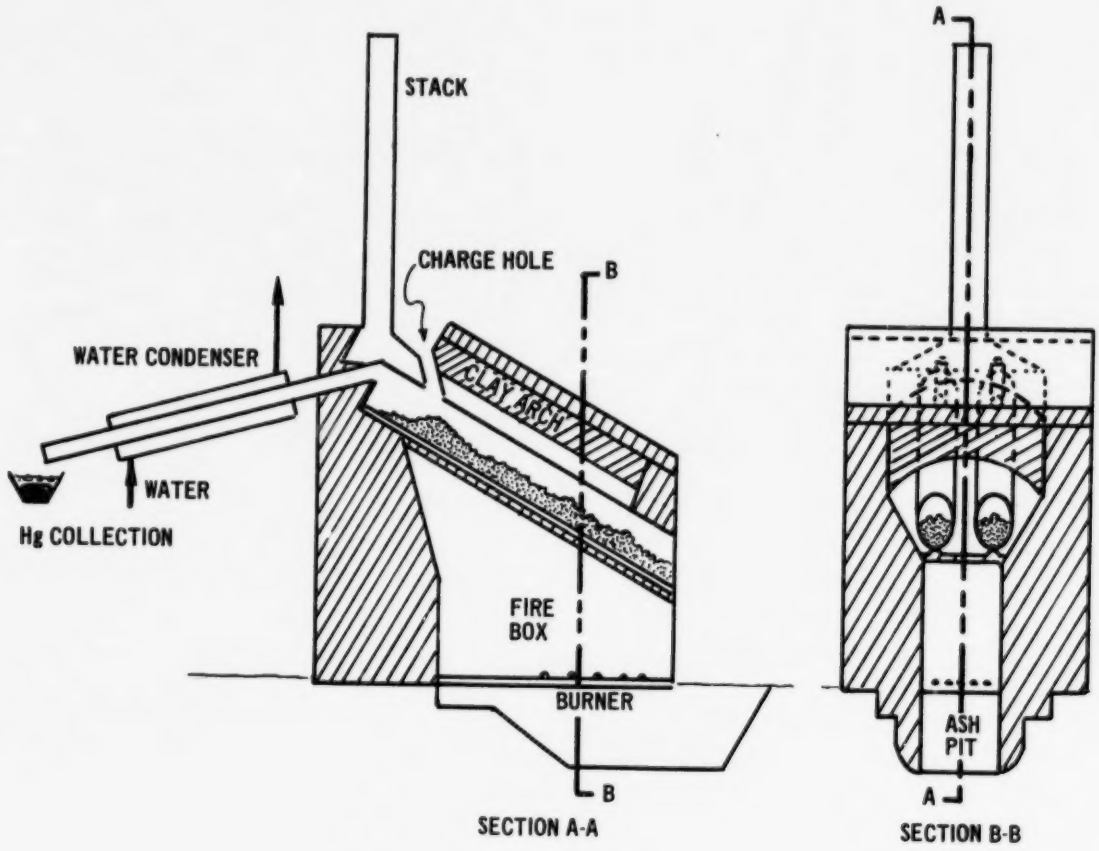

Figure 3-5. Two-pipe, inclined pipe retort.[16]

rate is low. Possibly the largest emission occurs during the opening and closing of the retort door for removal of spent ore and charging of ore concentrate.

Other emissions are from the retort room ventilation air, from the hoeroom or table ventilation air, and from the calcined concentrate. If good housekeeping practices are followed, these emissions can be minimized.

3.1.2 Emission Reduction Resulting from Process Changes

Reductions of mercury emissions can be accomplished by adding a control device to remove mercury from a gas or by using a process that inherently produces lower emissions. For new plants, a process that would produce less mercury emissions would probably be employed. For existing plants, a mercury control device might be added to the existing process to reduce emissions; however, in some cases, it may be more economical to convert to a different process. The decision as to which course of action to follow would probably be determined by an economic evaluation of both control methods. This section will discuss emission reductions resulting from process changes; Section 3.1.3 will discuss emission reductions resulting from the application of control techniques.

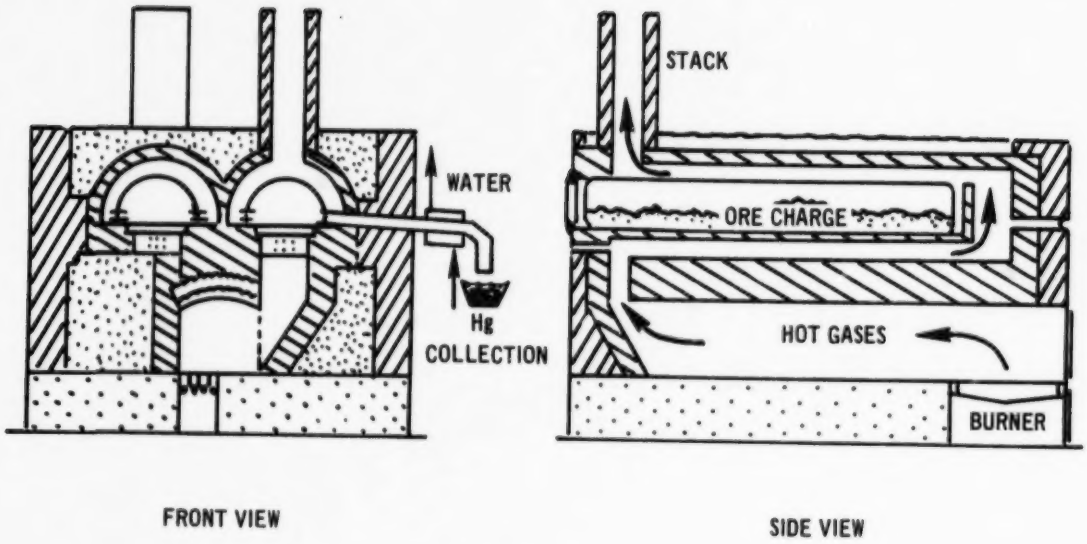

Figure 3-6. D - retort.[16]

3.1.2.1 Pyrometallurgical Processing

The largest mercury emission from a mercury extraction facility using a directly heated furnace occurs from condenser stack gas emissions. These gases are saturated with mercury vapor and also contain entrained particulates. The major parameters governing the emission rate are stack gas flow rate and temperature. If either or both of these parameters are reduced, the mercury emissions will also be reduced.

The temperature cannot generally be reduced by a process change alone; a control technique to cool the effluent would have to be employed in most situations. This technique will be discussed in Section 3.1.3. For the purposes of this discussion, the temperature will be assumed constant while the stack gas flow rate is reduced by different processes. A reduction in stack gas flow rate can be accomplished by converting from a direct- to an indirect-heating process or by beneficiation of ore prior to furnacing.

3.1.2.1.1 Indirect Heating – Indirect heating of ore, either concentrated or as mined, will effect a reduction in mercury emissions from the stack. The stack temperature generally determines the concentration of mercury in the stack gas; therefore, if the gas flow rate is reduced, a proportional emission reduction will occur at the same temperature. The retort process and the multiple-hearth furnace using muffle or electrical resistance heating can be used. These processes are described in Sections 3.1.1.2 and 3.1.1.4. The gases evolving from the ore during roasting would be the only gases produced and would consist of moisture, sulfur dioxide, air, mercury vapor, and small quantities of other gases. It is estimated that the stack gas flow could be reduced by 70 to 80 percent.

Because it is used for batch processing, the retort is generally used only on a small scale. The indirectly heated multiple-hearth furnace requires a larger hearth area for the same ore rate than the directly heated furnace requires because of the lower heat transfer efficiency inherent with indirect heating. The larger area requirement would increase the

capital investment and operating cost of an indirectly heated furnace by a sizeable factor.

3.1.2.1.2 Beneficiation – Ore grade is improved by the beneficiation processes of hand sorting, jigging, tabling, and flotation. As an example, if 100 tons of low-grade mercury ore were concentrated by beneficiation methods to 2 tons of higher-grade concentrate, the fuel needed to volatilize the mercury would be reduced since 98 tons of non-mercury-containing ore would not have to be treated from ambient temperature to 1200°F. The lower fuel requirement would result in a lower stack gas flow rate, thereby resulting in a sizeable reduction in emissions. The emission reduction factor is estimated to be roughly equal to the concentration ratio. In this example, the concentration ratio is 100/2, or 50. If both indirect heating and beneficiation were used in the same process, an even larger emission reduction would result.

3.1.2.2 Hydrometallurgical Processing

Hydrometallurgical mercury ore processing produces essentially no atmospheric mercury emissions. The only potential emission is that of ore dust resulting from crushing and grinding operations. Soluble mercury compounds probably remain in the tailing and leach solutions from the hydrometallurgical operation, and these may create a water pollution problem. Since virtually no data are available on potential losses of mercury in the solid residue and waste waters from sizeable hydrometallurgical operations, it would be difficult to estimate either the magnitude of the water pollution problem or the cost of alleviating it. Research and pilot studies have indicated that water pollution problems can be minimized if the operation is properly designed.

3.1.3 Control Techniques

The primary mercury industry currently employs little if any control technology for reducing mercury emissions from the condenser stack gases. As a result, few of the techniques that are discussed as control methods have been actually used in this industry. Cooling, mist elimination, water scrubbing, and the use of activated carbon are control methods that have been used successfully in reducing mercury emissions from similar gas streams and should be applicable to reducing emissions from primary mercury extraction plant effluents.

3.1.3.1 Directly Heated Furnaces

3.1.3.1.1 Cooling and Condensing – The basic technique currently employed for minimizing mercury emissions from the condenser stack is the control of the temperature of the effluent gases. A stack effluent temperature range of 90° to 110°F will allow from 5.2 to 12.4 pounds of vaporous mercury to be emitted to the atmosphere per day when the stack flow rate is 1600 standard cubic feet per minute. This is a typical stack flow rate for a 100-ton-per-day mercury extraction facility and will be used for subsequent examples. Assuming that the emission of particulate mercury at least equals the amount lost as vapor, the total loss of mercury from the condenser stack would range from 10.4 to 24.8 pounds per day.

As the effluent temperature is lowered, the mercury vapor content of the condenser gas stream is decreased. This temperature dependency is illustrated in Table 3-3 for five volumetric flow rates and an assumed original condenser outlet temperature of 110°F.

Cooling of the condenser gases can be accomplished by the use of either direct or indirect cooling techniques. Because of the large particulate loading of the condenser gas stream, direct cooling methods may possess an advantage over indirect methods since direct cooling aids in mist and particulate removal. The use of direct cooling, however, also introduces the necessity of water treatment facilities and creates the possibility of a water pollution problem.

If a supply of low-temperature water is available for use as a cooling medium, the cost of either a direct- or indirect-cooling system can be substantially reduced. The majority of

Table 3-3. CALCULATED VAPOROUS MERCURY EMISSIONS FOR SELECTED CONDENSER STACK FLOW RATES AND TEMPERATURES

Condenser volumetric flow rate, scfm[a]	Condenser exit temperature, °F	Final temperature difference, °F	Condenser exit Hg vapor concentration, mg/m^3 [a]	Hg vapor condensed by cooling, lb/day	Hg vapor atmospheric emission, lb/day
500	110	0	86.01	0	3.87
	80	30	23.40	2.81	1.06
	70	40	14.62	3.21	0.66
	60	50	9.02	3.47	0.40
	55	55	6.96	3.56	0.31
1500	110	0	86.01	0	11.61
	80	30	23.40	8.45	3.16
	70	40	14.62	9.64	1.97
	60	50	9.02	10.45	1.22
	55	55	6.96	10.67	0.94
2000	110	0	86.01	0	15.49
	80	30	23.40	11.28	4.21
	70	40	14.62	12.86	2.63
	60	50	9.02	13.87	1.62
	55	55	6.96	14.23	1.26
2500	110	0	86.01	0	19.39
	80	30	23.40	14.12	5.27
	70	40	14.62	16.10	3.29
	60	50	9.02	17.35	2.04
	55	55	6.96	17.82	1.57
3500	110	0	86.01	0	27.01
	80	30	23.40	19.66	7.35
	70	40	14.62	22.41	4.60
	60	50	9.02	24.16	2.85
	55	55	6.96	24.81	2.20

[a]Standard conditions, 70°F and 29.92 in. Hg.

the primary mercury extraction facilities do not, however, have access to naturally occurring low-temperature water, and some form of chilling is required. If water is available but is not of low enough temperature to cool the effluent adequately, a chilled brine or Freon refrigeration system could be employed to produce low-temperature water. At those sites where water is not available, a closed-loop refrigeration system could be used.

In order to be effective, the cooling step should be followed by some type of mist elimination device.

3.1.3.1.2 Mist Elimination – The condenser gas stream may contain mercury as mercury vapor, mercury oxides, mercury mists, and mercury adsorbed on soot and other particulate matter. Results of source tests by the Environmental Protection Agency have indicated that as much as 50 to 70 percent of

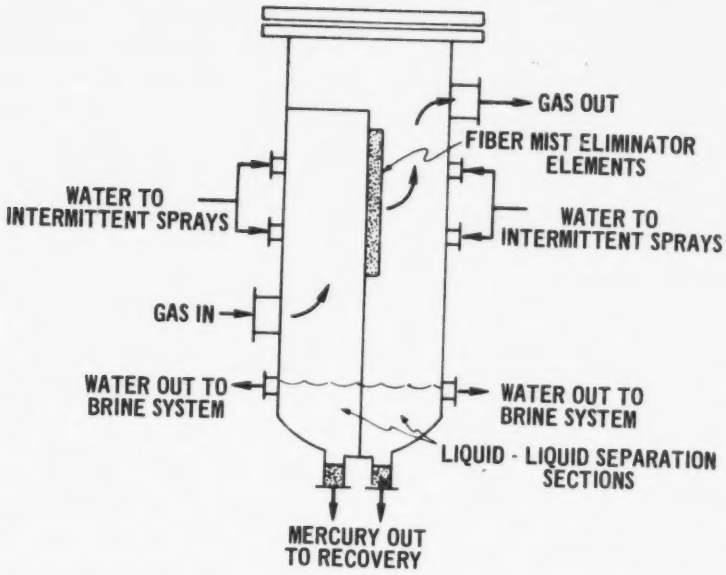

Figure 3-7. Mercury mist eliminator.[17]

the mercury that is emitted from the stacks of primary extraction facilities is in the particulate form.[9-11] Total mercury stack emissions, therefore, may be two to three times larger than the calculated vaporous emissions presented in Table 3-3.

In one type of commercially available mist eliminator, the mercury-particulate-laden gases pass horizontally through a fiber bed enclosed in screens. Clean gases exit from this fiber bed, whereas the separated mercury drains to the bottom of the mist eliminator. Inertial impaction, Brownian movement, and direct interception are the three basic mechanisms utilized for mercury mist separation. In order to increase the rate of drainage of the collected mercury from the fiber bed, water or other liquid is intermittently or continuously sprayed onto both sides of the bed. The lower section of the eliminator serves as a coalescer for the separation of the collected mercury from the water. Figure 3-7 illustrates this device.

Since mercury mist eliminators are currently applied in the treatment of relatively clean gas streams (that is, low particulate loading of contaminants other than mercury), the effectiveness of this device for extracting particulate mercury from a stream containing high amounts of particulate matter is not known. Additional particulate loading may cause the efficiency of this technique to be lower than normal. Use of additional flushing water within the mist eliminator or installation of air precleaning equipment prior to the mist elimination device may effectively eliminate the problem.

The particulate mercury removal efficiency of a mercury mist eliminator has been estimated at 86 percent.[18,19]

3.1.3.1.3 Wet Scrubbing – Wet scrubbing devices employ a variety of mechanisms to collect particulate matter. Interception of particulate matter by liquid droplets resulting in a heavier dust-liquid agglomerate is the most important of these mechanisms. A particle that has collided with a liquid droplet resists separation because of van der Waals forces. Particulate matter collected on a liquid droplet in this manner can be efficiently

removed from the gas stream by a centrifugal collector. The particulate matter collection efficiencies of these devices vary with energy input and can extend over a wide range depending on the design.

The liquid-solid slurry formed can be sent to a settling tank either directly from the device or from a subsequent centrifugal collector. The settled scrubbing liquid may be pumped back to the scrubber and reused. A bleed stream must be taken off of the settling tank to maintain a stable level in the tank since particulate matter and moisture are removed from the gas stream. The bleed stream usually represents 3 to 5 percent of the liquid-solid slurry and is a potential source of water pollution. This stream can be discharged into a secondary settling tank where the mercury solids are separated, and the liquid either discharged or given additional treatment.

Scrubbing systems have several advantages when compared to other particulate matter collection devices:

1. The required capital expenditure is normally lower for scrubbers than for other types of gas-cleaning equipment.
2. The collection efficiency of the scrubber is flexible, depending on the power input.
3. A large range of particulate sizes can be collected.
4. There are no secondary dust problems since the disposal of collected contaminants is a wet operation.
5. The cost of maintenance is low since there are few or no moving parts.
6. Scrubbers are compact in size.
7. Since the collecting operation is wet, there is virtually no limitation on the gas stream humidity and temperature.
8. Wet scrubbers allow the simultaneous collection of pollutants that are in the gaseous, liquid, or solid form.

There are numerous types of wet scrubbers in use that remove particulates from a gas stream, examples of which are the packed-bed scrubber and the venturi scrubber.

Several variations of low-energy, low-efficiency, packed-bed scrubbers have been employed in the primary mercury industry to remove mercury from gaseous streams. In one domestic primary extraction facility, a system has been used that consists of water sprays in a packed redwood tank.[20] Another extraction operation scrubs its condenser gases in a similar packed-bed water scrubber.[21] In most situations, the low-energy scrubbing system will prove inadequate for mercury removal because of its low particulate removal efficiency.

The venturi scrubber is a high-energy, high-efficiency scrubber that has been used to remove particulate matter from gas streams similar to the condenser stack gas streams of primary mercury extraction facilities. The water necessary to scrub a 1600-standard-cubic-foot-per-minute gas stream is estimated to be between 5 and 9 gallons per minute, depending on the gas loading and size distribution of the particulate matter. It is expected that a particulate mercury collection efficiency of 95 percent could be achieved with a pressure drop of 20 to 30 inches of water through the venturi scrubber and cyclone.[22] A suction fan located after the cyclone is necessary to overcome this pressure drop.

A venturi scrubber that produces sufficient turbulence within the venturi to allow cooling and condensing to occur is commercially available.[22] The scrubbing water required for this situation would necessitate cooling prior to injection. This type of venturi scrubber would not require precooling of the gas stream in order to achieve good collection efficiencies.

3.1.3.2 Control of Indirectly Heated Furnace Operations by Use of Treated Activated Carbon

The use of either sulfur- or iodine-impregnated activated carbon as a control technique for the removal of mercury vapor

from condenser gases of a retort operation should prove adequate. Some problems could arise if substantial amounts of particulate mercury are present in the gas stream; however, this situation can be corrected either by preheating the gas stream or by using a mist elimination device prior to the carbon bed. Although activated carbon has not been used specifically for the treatment of the condenser stream of an indirectly heated primary extraction facility, this technique has been successfully used to treat similar process streams. The mercury vapor is adsorbed by the carbon and reacts with the impregnated sulfur or iodine to form mercury compounds.

In order for treated activated carbon to perform efficiently, the gas stream velocity through the bed should be in the range of 20 to 40 feet per minute.[23] This low gas velocity is required to allow sufficient contact time between the mercury and the treated carbon. As previously mentioned, the particulate loading of the gas stream, including both particulate mercury and other solid and liquid contaminants, must be low or the carbon bed will plug and lose its efficiency. Destructive distillation of the spent carbon appears practical for recovering the adsorbed mercury.

Because of the low condenser gas flow rates and the low particulate loading in a retort operation, it is estimated that the mercury vapor collection efficiency of the treated activated carbon could approach 99 percent.[23]

3.1.4 Control Costs

Four basic methods for the removal of mercury from the condenser stack gases of a primary extraction facility have been described. These four techniques are cooling and condensing, mist elimination, wet scrubbing, and treated activated carbon. This section will present capital and annual operating costs for each of the preceding control techniques. All cost estimates are based on a model mercury extraction facility of 100 tons of ore per day capacity having a condenser stack gas flow of 1600 standard cubic feet per minute at 110°F. Equipment costs are based on the use of titanium and titanium-clad construction materials, which are required because of the corrosive nature of the condenser gases. All equipment costs were obtained directly from various users and vendors of control equipment.

The capital costs of specific systems are itemized and listed in the Appendix, Section A.3. The method employed for estimating the capital requirement and the annual cost is outlined in Section A.2.

3.1.4.1 Cooling and Mist Elimination

A control system that utilizes cooling to 55°F followed by partial mist elimination by means of a knockout drum could reduce emissions from the condenser stack gas stream to 5.8 pounds per day. If indirect cooling were used, the capital and annual operating costs for the preceding system would be $76,000 and $23,000, respectively. It is estimated that the use of a direct cooler would reduce the capital requirement to $51,000. The annual operating cost for this system would be $15,000. This estimate, however, does not include the cost of water-treatment facilities, which could be substantial.

The addition of a mist elimination device, similar to the one described in Section 3.1.3.1.2, to the preceding control system could reduce emissions to 1.7 pounds per day. The capital and annual operating costs for such a system utilizing indirect cooling would be $108,000 and $32,000.

Table A-5 of the Appendix provides more complete data on capital costs and expected emissions. Figure 3-8 illustrates a manufacturer's estimate of the purchase cost of a mist eliminator as a function of volumetric flow rate.

3.1.4.2 Wet Scrubbing

The cost of controlling atmospheric mercury emissions by means of a scrubbing technique will vary considerably with the type of scrubbing system employed.

The cost of a low-energy scrubber

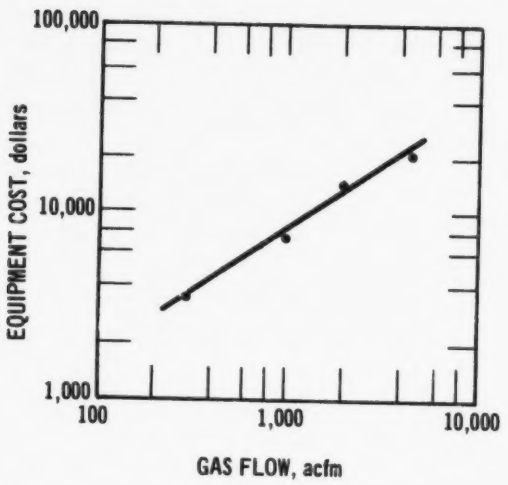

Figure 3-8. Manufacturer's estimate of purchase cost of a fiber pad type of mist eliminator.

(wooden tank) similar to the type employed by one domestic extraction plant is minimal. It is estimated that the capital requirement would be approximately $2000. This cost does not allow for a chiller or for water purification facilities; moreover, the effectiveness of this system is questionable.

The estimated equipment cost of a packed scrubbing tower constructed of carbon steel is about $0.30 per cubic foot per minute of gas flow.[24] Adjusting this price for the use of a corrosion-resistant construction material, such as fiber glass, and adding the cost of a water pump and a blower to handle a pressure drop of 3 inches of water, the capital requirement for a packed tower of 1600 standard-cubic-foot-per-minute capacity would be $5000. The capital and operating costs for a control system utilizing a packed tower and chilled water for direct cooling of the condenser gases to 55°F have been presented previously in Section 3.1.4.1. Insufficient data are available with which to evaluate the efficiency of a packed tower for the removal of mercury particulate.

The use of a venturi scrubber for removal of mercury particulate has been discussed in Section 3.1.3.1.3. It is estimated that a control system using an indirect cooler to cool the condenser gas stream to 55°F followed by a venturi scrubber could reduce emissions to 1.7 pounds per day. The capital cost for this system would be $86,000. The annual operating cost is estimated to be $26,000. These costs assume a capital cost of $12,000 for a standard venturi scrubber.

A second type of venturi scrubber, which has the ability to cool the gas stream and scrub simultaneously, can be purchased as a complete unit for approximately $14,000 for a facility treating 100 tons of ore per day. The only additional costs that would be incurred are for cooling the scrubbing water and for two adequately sized settling tanks for the scrubber discharge and the bleed stream. The manufacturer of this system considers that the packaging of this system will allow a minimal installation cost. The capital requirement for the complete system is estimated to be $30,000. No information is presently available to enable estimation of the degree of cooling that can be attained.

3.1.4.3 Treated Activated Carbon

The use of treated activated carbon as a control technique has been discussed in Section 3.1.3.2. To be effective, this system must be preceded by some type of mist elimination device. A carbon bed system preceded by a venturi scrubber could reduce the stack gas emissions to 1.8 pounds per day for a 100-ton-per-day facility. (This estimate is based on a conservative carbon bed vapor collection efficiency of 90 percent.) The expected capital and annual operating costs are estimated to be $66,000 and $20,000. More complete cost data are presented in Table A-5 of the Appendix.

3.1.4.4 Summary

Several control techniques and their respective costs have been discussed in Sections 3.1.3 and 3.1.4. Table 3-4 presents a summary

Table 3-4. SUMMARY OF PRIMARY MERCURY CONTROL TECHNIQUE COSTS AND EXPECTED EMISSIONS IN CONDENSER STACK GAS STREAM FOR 100-TON/DAY FACILITY

Control system	Capital cost, $	Annual operating cost, $	Expected emissions, lb/day
Cooling and mist elimination (Section 3.1.4.1 and Tables A-4 and A-5)	76,000 to 108,000	23,000 to 32,000	5.8 to 1.7
Wet scrubbing (Section 3.1.4.2 and Tables A-4 and A-5)	86,000	26,000	1.7
Treated activated carbon (Section 3.1.4.3 and Tables A-4 and A-5)	66,000	20,000	1.8

of both capital and operating costs for these techniques together with expected emissions. The capital costs of specific control systems are itemized in Table A-5.

All costs have been based on a facility of 100 tons of ore per day capacity. Cost estimates can be adjusted for other capacities by using the following equation:

$$C_A = C_{100} \left(\frac{P_A}{100 \text{ tons/day}} \right)^{0.6}$$

where: C_A = applicable control costs, equipment or capital

C_{100} = control cost for a 100-ton-per-day facility, equipment or capital

P_A = applicable extraction facility capacity in tons of ore per day.

3.1.5 Development of New Technology

This section contains information concerning processes and control techniques that are not considered to be "state-of-the-art" in the primary mercury extraction industry. Included in this section are processes and techniques that have been tested at the research and/or development levels but that have not been sufficiently demonstrated at full scale.

3.1.5.1 Hydrometallurgical Processing

3.1.5.1.1 Process Description – Over 90 years ago, Volhard reported that an aqueous solution of sodium sulfide could be used for dissolving cinnabar. Until recently, however, the only hydrometallurgical technique in use was that developed in 1915 for the recovery of mercury from amalgams. In the late 1950's, the U.S. Bureau of Mines conducted considerable research on the use of sodium sulfide and sodium sulfide-sodium hydroxide solutions for leaching mercury ores and concentrates.[25-27] These investigations demonstrated that cinnabar could be dissolved in these solutions.

The great variations in composition of ores, the cost of grinding ores finely enough for effective leaching, the simplicity and efficiency of pyrometallurgical processing, and the cost of reagents have generally precluded the consideration of hydrometallurgical processes for the recovery of mercury. However, if flotation is used to concentrate the ore, the cost of alkaline sulfide leaching followed by electrolytic precipitation of the mercury has been estimated to be about the same as the cost of pyrometallurgical treatment.

In 1970, U.S. Bureau of Mines investigators reported that recoveries of 90 to 99 percent could be attained by electro-

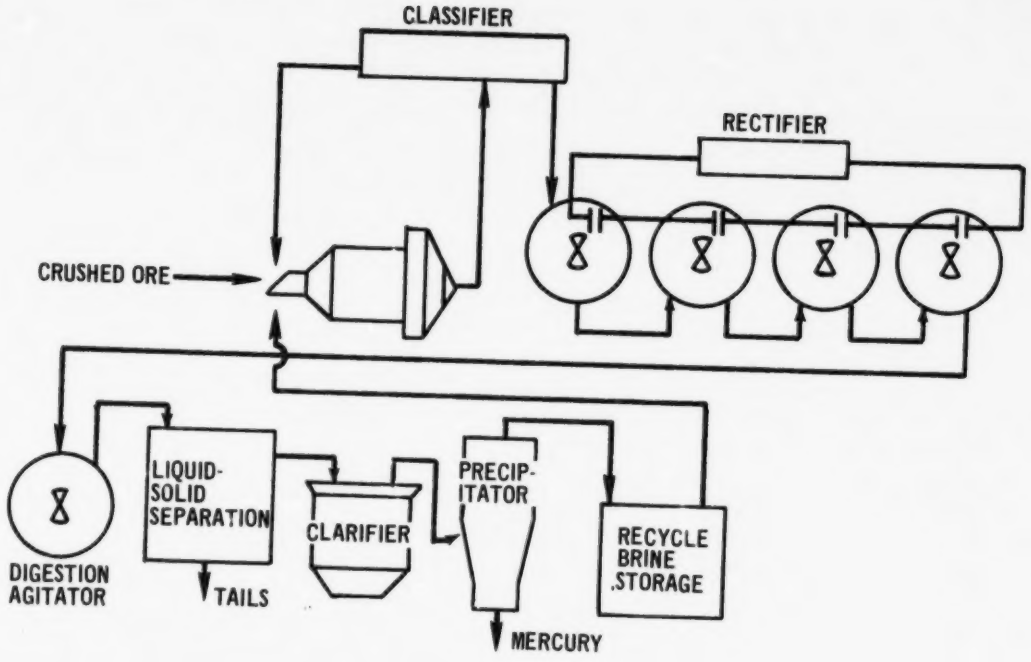

Figure 3-9. Conceptual hydrometallurgical plant layout and flow diagram based on laboratory data.[28]

oxidation of finely ground mercury ore in a brine solution to form soluble mercuric salts, followed by precipitation of the mercury with an active metal dust such as zinc, iron, or aluminum.[28] The process is applicable to low-grade mercury ores, and beneficiation is not necessary. The most important parameters in the electro-oxidation process are temperature, salt concentration, current density, type of electrodes, electrode spacing, treatment rate (amperes per ton of ore), and particle size of ore. The ore must be ground to 35 mesh or finer. In typical laboratory experiments, 1 to 7 hours of electrolysis was required at a 35 percent pulp density in a brine solution containing 4 to 20 percent sodium chloride.[28] Power consumption ranged from 10 to 50 kilowatt-hours per ton of dry ore.

The tailings are discharged into a settling pond. The concentration of mercury in the tails ore is approximately 0.1 pound per ton of solid tailing.[13] The concentration of mercury in the tails solution is approximately 1 part per million.[13] Figure 3-9 shows a conceptual plant layout and flow diagram based on laboratory data.

The operating cost for the electro-oxidation process is estimated to be $2 to $3 per ton compared to $5 to $8 per ton for a furnacing operation.[13] A feasibility study of this process conducted by a large engineering firm indicated that it was favorable for low-grade ores in the 2- to 3-pound mercury per ton range.[13] The U.S. Bureau of Mines is conducting pilot mill experiments in a 100- to 200-pound-per-hour pilot plant to quantify power and reagent requirements. A report is being prepared to give final details of this research project.

3.1.5.1.2 Emissions – The use of hydrometallurgical techniques for treating mercury ores would minimize or eliminate the emission of mercury to the atmosphere if the processing

Table 3-5. CHLORINE CAPACITY AND PRODUCTION METHODS IN THE UNITED STATES[31,32]

Year	Capacity, short tons/day	Percent of total installed capacity		
		Diaphragm cells	Mercury cells	Fused salt and nonelectrolytic
1946	4,012	88.6	4.3	7.1
1956	10,300	81.6	12.4	6.0
1962	14,697	76.2	18.5	5.3
1963	15,503	74.1	20.8	5.1
1964	16,404	72.2	23.0	4.8
1965	17,245	71.2	24.2	4.6
1966	18,939	69.7	26.5	3.8
1967	21,216	69.8	26.7	3.5
1968	23,238	68.1	28.6	3.3
1969	25,124	69.2	27.9	2.9
1970	28,276	69.6	27.2	3.2
1971	29,131[a]	69.8	27.2	3.0

[a] Preliminary.

plant were properly designed. To avoid the emission of finely divided ore particulates, the crushing and grinding circuit must provide maximum control of the dust. It is probable that soluble mercury compounds would remain in the tailings from the leaching operation even with careful countercurrent washing. The potential water pollution problem created could be minimized or eliminated by a properly designed tailings disposal system.

3.1.5.2 Sulfuric Acid Scrubber

A foreign concern has used a sulfuric acid scrubber to remove mercury and selenium from its nonferrous smelter sulfur dioxide gas stream.[29,30] This concern states that this system can reduce the mercury concentration from about 60 milligrams per cubic meter to about 200 micrograms per cubic meter in a 29,000 cubic-foot-per-minute gas stream. The cost of this sulfuric acid scrubber is approximately $500,000.

3.2 CHLOR-ALKALI PRODUCTION

3.2.1 History

Chlorine is produced almost entirely by electrolytic methods from fused chlorides or aqueous solutions of alkali-metal chlorides. In the electrolysis of an aqueous solution of potassium or sodium chloride, chlorine is produced at the anode while hydrogen and either potassium hydroxide or sodium hydroxide are produced as a result of processes occurring at the cathode. This requires that the anode and cathode products be kept entirely separate. Consequently, many ingenious cell designs have been developed and refined; all of these have been variations either on the diaphragm cell or on a cell which employs mercury metal as an intermediate cathode. Historically, these two processes were developed more or less in parallel. In the United States, the mercury process was an early leader but diminished to less than 5 percent of the installed chlorine capacity by 1946. From 1946 to 1968, the use of the mercury cell grew toward 28 percent of the total installed U.S. chlorine capacity. Since 1968, there has been a slight negative trend in the use of mercury cells. In Europe and most other parts of the world, the use of the mercury cell predominates. Detailed data on chlorine production by various production techniques are given in Table 3-5 and shown graphically in Figures 3-10 and 3-11. A listing of the operating mercury-cell plants in the U.S. is presented in Appendix A.4.

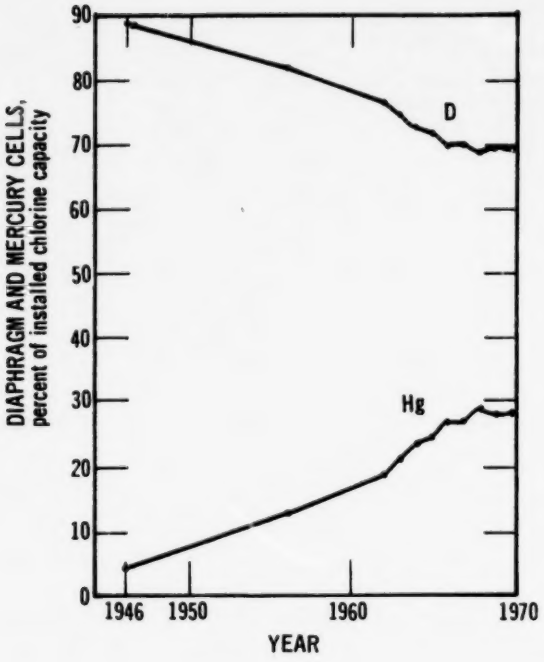

Figure 3-10. Percentage of total installed U.S. chlorine capacity for diaphragm and mercury cells.[31, 32]

3.2.2 Emissions and Process Description

3.2.2.1 Mercury-Cell Process

3.2.2.1.1 Process Description – Since the treatment of potassium chloride brines is essentially analogous to that of sodium chloride brines, the latter will serve as the basis for the following discussion. The basic process flow sheet for the production of chlorine and caustic soda is shown in Figure 3-12.

Purified and nearly saturated brine is fed continuously from the main brine treatment section into the inlet end of the electrolyzer, where it flows between a stationary graphite (or metal) anode and a flowing mercury cathode. The inlet end-box is a receptacle which is placed on the inlet end of the electrolyzer to provide a convenient connection for the stripped mercury as it returns from the decomposer (denuder). It also serves to keep the incoming mercury covered with an aqueous layer. The spent brine is recycled from the electrolyzer to the main brine treatment section of the plant; it may also be sent through a dechlorination step.

The chlorine gas product formed at the anode is discharged from the electrolyzer for further treatment. After cooling, the gas is dried by scrubbing with concentrated sulfuric acid. The spent acid from this drying step contains most of the trace amounts of mercury carried along by the wet chlorine gas. After compression, the dry chlorine may be used directly or may be subjected to a liquefaction step.

The sodium amalgam flows continuously from the electrolyzer through the outlet end-box to the decomposer; there it is the anode to a short-circuited graphite or metal cathode in an electrolyte of sodium hydroxide solution. The outlet end-box is a receptacle, which is placed on the outlet of the electrolyzer to provide a convenient means for keeping the sodium amalgam covered with an aqueous layer. The outlet end-box also permits the physical separation of these two streams. Purified water is fed continuously to the decomposer. This water reacts with the

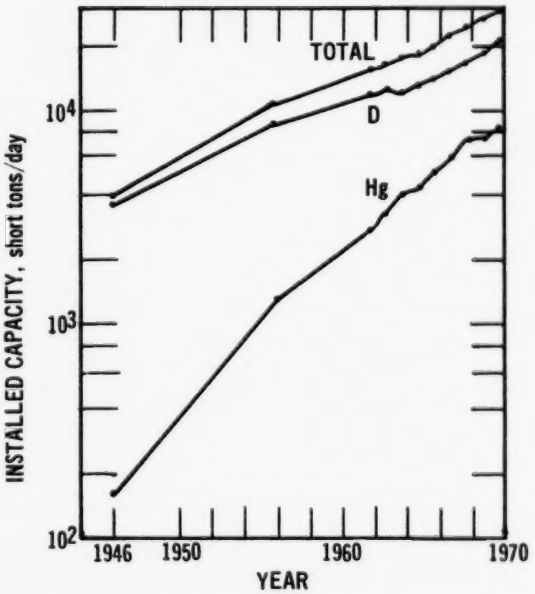

Figure 3-11. Installed U.S. chlorine capacity in diaphragm and mercury cells.[31, 32]

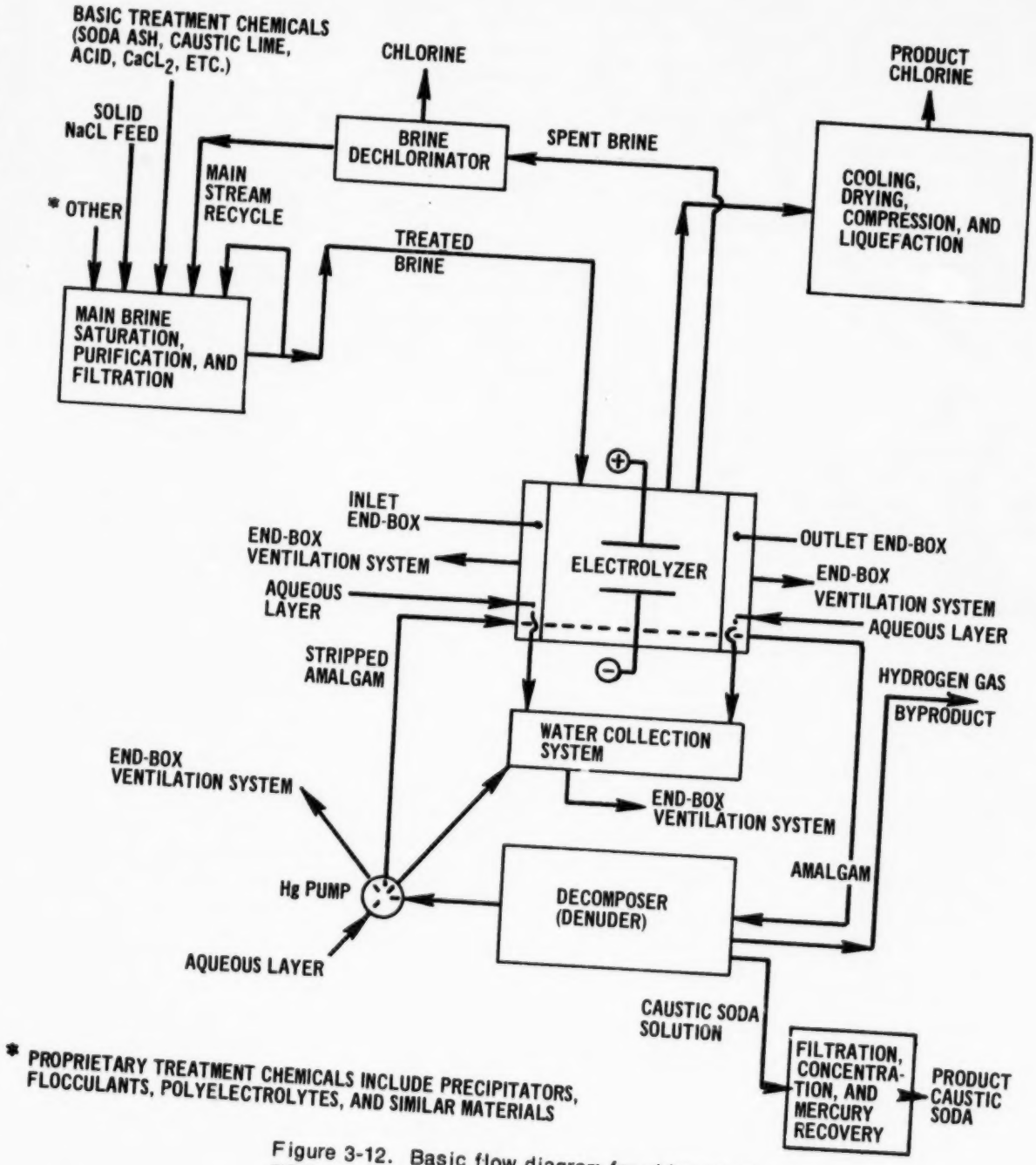

Figure 3-12. Basic flow diagram for chlor-alkali mercury-cell operation.[8]

sodium amalgam and produces sodium hydroxide solution as well as by-product hydrogen gas. About 800 to 850 standard cubic feet per minute of hydrogen is produced for each 100 tons of daily chlorine capacity. The high-purity caustic soda generally leaves the decomposer at a concentration of 50 percent sodium hydroxide by weight.

The caustic soda solution from the decomposer is usually sent to a filtration unit. The solid waste material from the filter may be processed to recover the mercury content in a retort. The mercury recovered from this retort operation is returned to the cell for reuse.

Filtered caustic solution at a concentration of 50 percent by weight may be further concentrated by evaporation to a 73 percent by weight sodium hydroxide product. In some instances, this material is heated to drive off the remaining water in order to produce anhydrous pellets or flakes of solid sodium hydroxide.

The by-product hydrogen gas from the decomposer may be vented to the atmosphere, burned as fuel, or used as a feed material for subsequent processing. This process stream is saturated with mercury vapor.

In order to reduce mercury vapor emissions to the cell room, an end-box ventilation system applies suction to various sections of the mercury cell operation. Such sections are usually one or both end-boxes, the mercury pump sump, and their water collection systems.

A large quantity of ventilation air maintains the temperature and mercury vapor concentration of the cell room at allowable levels.

3.2.2.1.2 Emissions – In terms of direct emissions of mercury to the atmosphere, the major sources are:

1. The hydrogen by-product stream.
2. The end-box ventilation system.
3. The cell room ventilation air.

The hydrogen by-product stream leaving the decomposer is saturated with mercury vapor (2.3 grams of mercury vapor per cubic meter at 210°F). If this stream were directly discharged to the atmosphere without prior cooling, an estimated 220 pounds of mercury would be emitted for each 100 tons of chlorine produced.

The minimum treatment known to be used consists of cooling the hydrogen stream to 110°F and partial mist elimination. For hydrogen saturated with mercury vapor at this temperature, the daily loss from the stream following treatment would be 6.8 pounds of mercury vapor per 100 tons of chlorine produced. The entrainment of condensed mercury in the hydrogen stream will result in emissions in excess of the above amount, which was calculated from known values of the vapor pressure of mercury. At least 86 percent of this additional loss of mercury as mist can be eliminated in a properly designed mercury entrainment separator.[18,19] The calculated combined emission of mercury vapor and mercury mist, after minimum treatment has occurred, is estimated to be 50 pounds for each 100 tons of chlorine produced.

Mercury and mercury compounds are collected from the end-boxes, the mercury pump sumps, and their water collection systems by the end-box ventilation system. Mercury emissions from this system are dependent on the percentage of mercury saturation and the volumetric flow rate. The volume, the degree of mercury saturation, and, in turn, the resulting emissions from the end-box ventilation system inherently depend upon the age of the plant, upon the type and specific configuration of the cells, end-boxes, and decomposers, and upon the standard operating procedures employed at a particular location. For some new cell modifications currently being installed, the volume of the end-box ventilation system is small compared with that of the hydrogen stream. In most cases, however, the volumetric flow rate of the end-box ventilation system may approach or exceed the flow rate of the hydrogen stream.[8,19,33] For the purposes of this discussion and subsequent emission and cost

calculations, the volumetric flow rate of this system will be assumed to be equal to that of the hydrogen stream. Preliminary results of Environmental Protection Agency testing indicate that the mercury emissions from an untreated or inadequately treated end-box ventilation system range from 2 to 15 pounds for each 100 tons of chlorine produced.[19,33,34]

The cell room ventilation system serves two major purposes. The primary function of this air stream is to cool the cell room environment, but it also provides a means of reducing the cell room concentration to within the recommended Threshold Limit Value (TLV). The Threshold Limit Value of the American Conference of Governmental Industrial Hygienists has recently been reduced from 100 micrograms per cubic meter to 50 micrograms per cubic meter.

The volumetric flow rate of the cell room ventilation stream varies from 100,000 to 1,000,000 cubic feet per minute for each 100 tons of daily chlorine capacity. A higher flow of air may be needed in old plants, where high mercury concentrations are more likely to exist. On the basis of data obtained from operating plants, it has been estimated that mercury emissions from the cell room ventilation system vary from 0.5 to 5.0 pounds per day per 100 tons of daily chlorine capacity.[8,19]

These emissions were calculated using the TLV of 100 micrograms per cubic meter. Figure 3-13 illustrates the daily mercury emissions as a function of cell room volumetric flow rate when a plant maintains its cell room air at a TLV of 50 micrograms per cubic meter and at 80 percent of this TLV, or 40 micrograms per cubic meter.

Mercury enters the cell room atmosphere as a result of a number of operations or conditions, a few of which are listed below:

1. End-box sampling.
2. Removal of mercury butter from the end-boxes.
3. Cell maintenance and rebuilding operations.
4. Other maintenance work which exposes the internal surfaces of pipes and equipment.
5. Accidental spills of mercury.
6. Leaks from cells and mercury pumps.
7. Cell failure and other unusual circumstances.

The number and variety of sources of mercury emissions to the cell room air clearly indicate that careful plant operation and good housekeeping are essential in order to minimize the amount of mercury emitted into the cell room air. In Section 3.2.4.6, a list of housekeeping practices for minimizing mercury emissions to the cell room will be recommended.

3.2.2.2 Diaphragm-Cell Process

3.2.2.2.1 Process Description – Chlorine and caustic soda can be produced by the electrolysis of brine in a diaphragm cell. In this process, an asbestos diaphragm separates the anode from the cathode. Chlorine gas is formed at the anode and hydrogen gas and caustic are formed at the cathode. One disadvantage of this process is that the caustic is of a lower grade than that produced by the

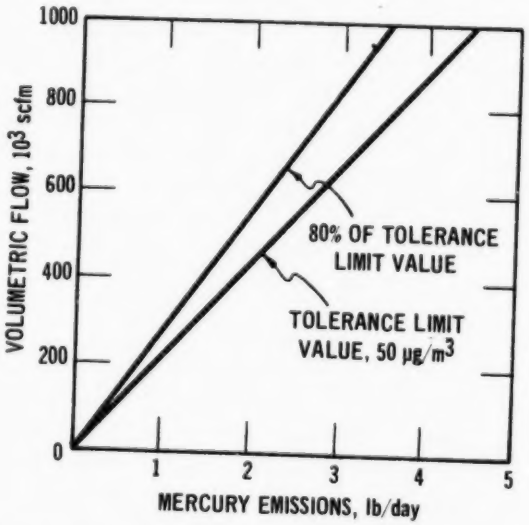

Figure 3-13. Mercury emissions in cell room ventilation air.

mercury cell and must be concentrated and purified for some uses.

3.2.2.2.2 Emissions – The diaphragm cell produces no mercury emissions.

3.2.3 Emission Reduction Resulting from Changing to Diaphragm-Cell Process

Reduction of mercury emissions can be accomplished by adding a control device to remove mercury from a gas stream or by converting to a process that inherently produces lower emissions of mercury. This section will discuss process changes; Section 3.2.4 will discuss emission reductions resulting from the application of control techniques.

Replacement of the mercury-cell chlor-alkali plant with a diaphragm-cell plant is an effective, but expensive, method for eliminating all mercury emissions from the manufacture of chlorine and sodium hydroxide.

The diaphragm-cell chlor-alkali plant produces chlorine at a cost usually slightly less than that of the mercury-cell chlor-alkali plant, but the sodium hydroxide produced by the diaphragm cell is only about 11 percent sodium hydroxide by weight and is saturated with sodium chloride. The sodium hydroxide produced by the mercury cell is about 50 percent sodium hydroxide by weight. In order for the caustic produced by the diaphragm cell to be competitive with the mercury-cell caustic, it must be upgraded. The process for upgrading weak caustic from diaphragm-cell chlor-alkali plants, while somewhat expensive to install and operate, has been developed for some time and has recently been improved. The 11 percent sodium hydroxide from diaphragm plants can be concentrated to 50 percent in multiple-effect nickel evaporators to obtain a 50 percent sodium hydroxide solution with 1 percent sodium chloride content.[35] This material is somewhat turbid when compared to 50 percent sodium hydroxide from mercury-cell chlor-alkali plants.

Impurities, such as sodium chloride and trace metals, are often contained in the concentrated diaphragm-cell caustic. Since these impurities cannot be tolerated by certain industries requiring high-purity caustic for processing, a proprietary process, which reduces these impurities, has been developed and is available for licensing. The process, known as the DH process, utilizes anhydrous ammonia in a liquid-liquid extraction operation to reduce the sodium chloride concentration in 50 percent diaphragm-cell caustic solution from 1.0 to 0.025 percent.[35] This grade of 50 percent diaphragm-cell caustic is nearly as pure as the 50 percent caustic produced from mercury cells and should be able to meet all or nearly all of the market for a high-grade 50 percent sodium hydroxide. The cost of producing the diaphragm-cell caustic is somewhat higher than that of the mercury-cell caustic. The costs of conversion will be discussed in Section 3.2.5.1.

3.2.4 Control Techniques

Various techniques have been developed for the control of mercury emissions from chlor-alkali operations. These techniques apply to the by-product hydrogen stream and the end-box ventilation system.

3.2.4.1 Cooling and Condensing

3.2.4.1.1 Hydrogen Gas Stream – Hydrogen leaves the decomposer at 200° to 260°F and passes into the primary cooler, where ambient water is normally used in a shell-and-tube heat exchanger to cool this stream to 90° to 110°F (Figure 3-14). The primary cooler usually has a device, known as a mercury knockout drum, for separating condensed mercury from the hydrogen stream. After passing through this initial drum, the hydrogen stream may be subjected to additional cooling in a secondary cooler that uses chilled water or brine as the cooling medium. The condensed mercury from this secondary cooling step is partially removed in a second knockout drum. The temperature of the hydrogen stream discharged from the secondary cooler ranges from 37° to 55°F. Because of the rapid variation of mercury saturation

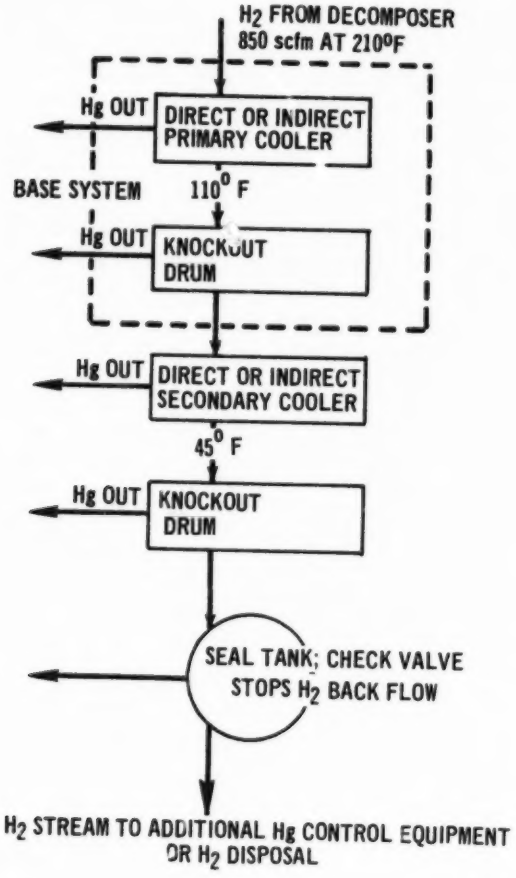

Figure 3-14. Cooling and condensing of hydrogen stream.

concentration with temperature, the hydrogen stream temperature should be decreased as much as possible in order to condense the largest amount of mercury vapor.

A large percentage of the condensed mercury may remain in the gas stream as a mercury mist that is difficult to efficiently separate from the hydrogen stream. One approach to resolving this problem is the use of a direct-contact cooler with chilled water or brine instead of a shell-and-tube heat exchanger. The chilled aqueous medium is often in a closed loop with a shell-and-tube heat exchanger that uses mechanical refrigeration. The bleed-off liquor from the direct-contact cooling system is sometimes treated with chemicals such as sodium hydrosulfide to remove the mercury; the clean liquor is then recycled back into the closed loop. An alternate method of treatment is to pump the bleed-off stream into a waste-water settling pond. The use of a direct-contact cooler has the disadvantage of requiring a water treatment system.

3.2.4.1.2 End-box Ventilation System – The temperature of the end-box ventilation air ranges from 160° to 180°F, and the air may be 10 to 50 percent saturated with mercury and nearly saturated with water. For calculation purposes, however, it is assumed that both the end-box stream and the hydrogen stream are saturated with mercury vapor. The maximum mercury content of the process streams is therefore considered. Some end-box ventilation gases contain enough chlorine to form mercuric chloride, which remains in the gas stream as particulate matter. The techniques used for cooling the end-box ventilation air are similar to the methods used to cool the hydrogen stream. However, the presence of mercuric chloride in this gas stream has led to a greater usage of direct-contact coolers for cooling and particulate matter removal. The high water content of this system has limited the temperature to which this gas stream can be cooled due to the formation of ice crystals. The presence of chlorine and mercury salt contaminants has also required the use of more corrosion-resistant construction materials such as titanium and titanium-clad steel.

A technique used in industry to treat end-box ventilation air is to cool the air stream with a direct-contact packed tower employing water as the coolant. This tower is followed by an indirect cooler, which uses 40° to 68°F cooling water. The resulting entrained particulate mercury is removed by a mist eliminator.[33] A mercury emission rate of 1 pound per day from the end-box ventilation system for each 100 tons per day of chlorine capacity has been estimated for such a system.

A considerable reduction in the capital and operating costs of the end-box system can be obtained by the reduction of the end-box ventilation flow rate. This reduction can be accomplished by the installation of leak-tight covers on all cell end-boxes and by the replacement of submerged mercury pumps with in-line pumps.

3.2.4.2 Mist Elimination

There are two basic types of mist eliminators that are commercially available to remove mercury mist from gaseous streams. One of these, which consists of fiber pads to remove entrained mist by mechanisms of impaction, interception, and Brownian movement, is discussed in Section 3.1.3.1.2 and shown in Figure 3-7. When this type of eliminator is used for mercury control in an end-box ventilation gas stream, backwashing with water or a strong reducing agent may become necessary to prevent plugging.

Another type of mist eliminator which is being used in at least one application utilizes a converging-diverging nozzle arranged so that the gases being cleaned follow a curved path and are acted upon by high centrifugal forces in the throat area (Figure 3-15).[36] These forces are reported to cause the coalescence of mercury mist and entrapment of submicron mercury mist upon the upper wall of the divergent section. Separated particles are washed away from the walls by sprayed liquid. This mist eliminator is reported to have an efficiency comparable to that of the fiber pad type of mist eliminator. The entire unit can be made of plastic or special alloys which resist amalgamation. Figure 3-16 presents a typical control system utilizing a mist elimination device which is applicable to either the hydrogen or the end-box streams.

3.2.4.3 Chemical Scrubbing Techniques

3.2.4.3.1 Depleted Brine Scrubbing System – Depleted brine scrubbing techniques have been applied for mercury removal from hydrogen and end-box ventilation gas streams in only a few instances. The depleted brine scrubbing technique uses the brine discharged from a chlorine cell as a scrubbing liquor. This depleted brine contains about 250 grams per liter of sodium chloride and 0.6 to 0.9 gram per liter of available chlorine; it has a pH of 2 to 4.[37] This solution is used as the scrubbing medium in a sieve plate tower or in a packed-bed scrubber. Upon contact with the brine scrubbing solution, mercury vapor and mist form soluble mercury complexes. The

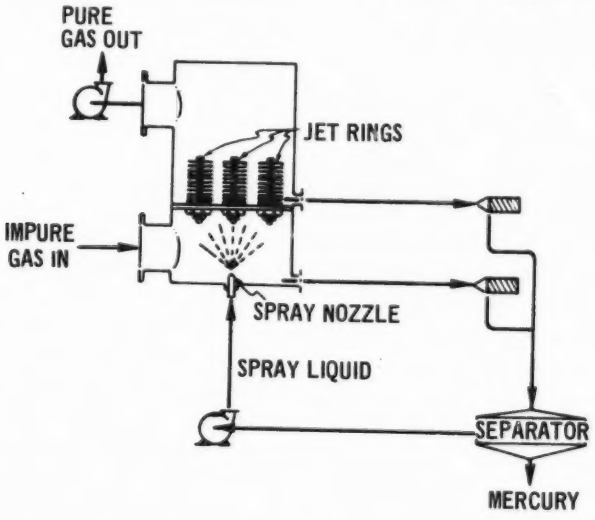

Figure 3-15. Converging-diverging nozzle mist eliminator.[36]

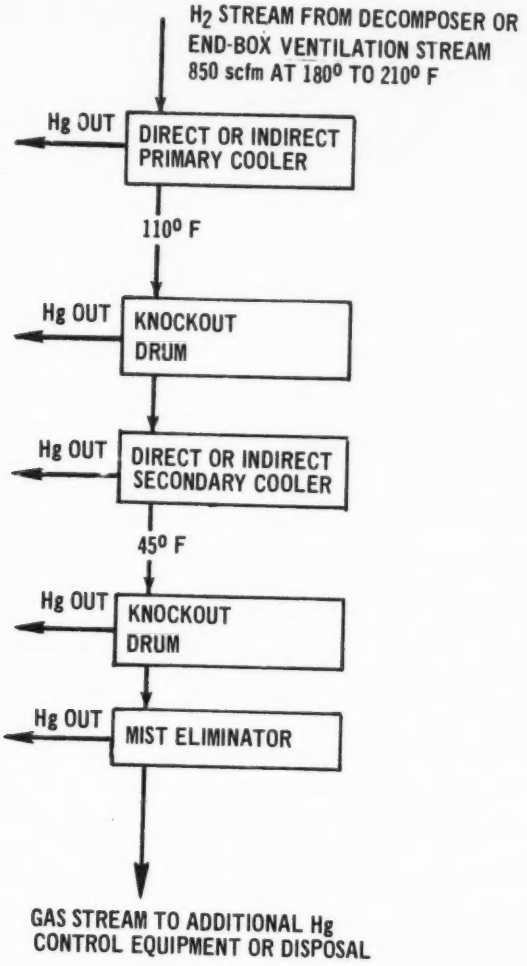

Figure 3-16. Cooling, condensing, and mist elimination.

mercury is subsequently recovered by electrolysis when the scrubbing medium is returned to the mercury cell. Figure 3-17 presents a simplified flow sheet for a depleted brine scrubbing system.

One application of a depleted brine scrubbing system (System A) has been used since the early 1960's for the removal of mercury from the hydrogen stream. The mercury concentration of the treated hydrogen has been reported to be approximately 85 micrograms per cubic meter. Mercury losses from the treated hydrogen stream would, therefore, be less than 0.01 pound per day on a 100-ton-per-day chlorine basis. In this system, the hydrogen gas is discharged from the decomposer at 180°F. It is then pumped into a direct-contact water scrubber where it is cooled to 100°F. After partial separation of the entrained mercury mist with a knockout drum, the hydrogen is cooled to 55°F by a direct-contact cooler using chilled brine in a closed-loop system. The cooled hydrogen

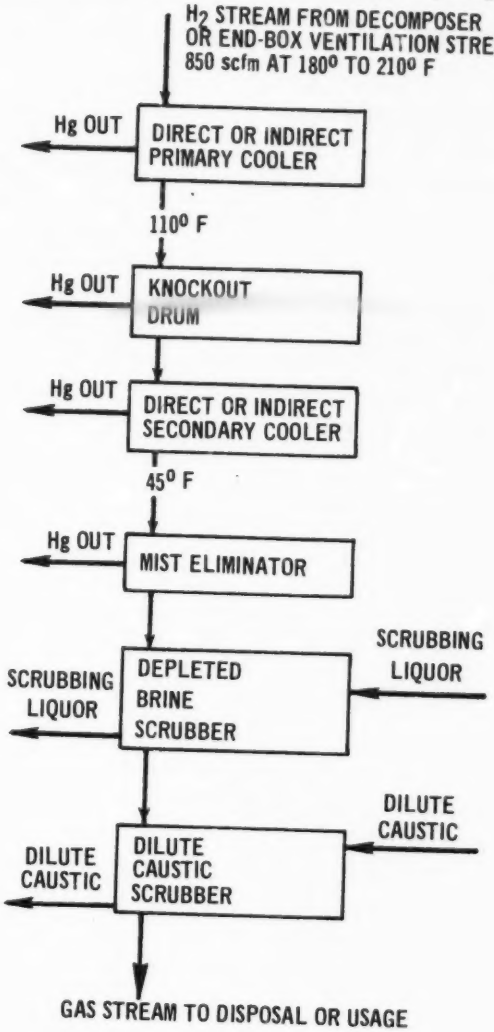

Figure 3-17. Depleted brine scrubbing system.

then flows through a mist eliminator and a depleted brine scrubbing tower. A final alkaline scrubber is used to remove entrained chlorine and acid.

A second application of depleted brine scrubbing utilizes a system similar to the one discussed above, but deletes the alkaline scrubber (System B). With this system, mercury emissions from the combined process streams are reported to be about 0.6 to 0.9 pound of mercury per day on a 100-ton-per-day chlorine basis.

3.2.4.3.2 Hypochlorite Scrubbing System – A second type of chemical scrubbing technique, which has been recently developed, uses a sodium hypochlorite solution as a scrubbing liquor. In one application of this technique, the scrubbing liquor consisted of equal molar amounts of sodium chloride and sodium hypochlorite. The scrubbing medium reportedly required a narrow pH control for optimum mercury removal efficiency. This narrow range of pH was difficult to maintain, and as a result, the sodium hypochlorite scrubber was converted to a depleted brine scrubber.

A second application of a sodium hypochlorite scrubbing technique, which is reported to have solved the problem of pH control, is available for licensing.[38] This system employs a dilute solution of sodium hypochlorite with a large excess of sodium chloride over the stoichiometric quantity. The mercury removal efficiency of this system is maintained over a wide enough pH range to make control possible.[38] This system has been employed successfully at two sites to date. A mercury collection efficiency of 95 to 99 percent has been reported.[39]

Figure 3-18 presents a general schematic flow sheet of the hydrogen stream for a control system using a hypochlorite scrubber. It is estimated that mercury emissions for the combined hydrogen and end-box streams would range from 0.2 to 0.8 pound per day for a 100-ton-per-day plant.

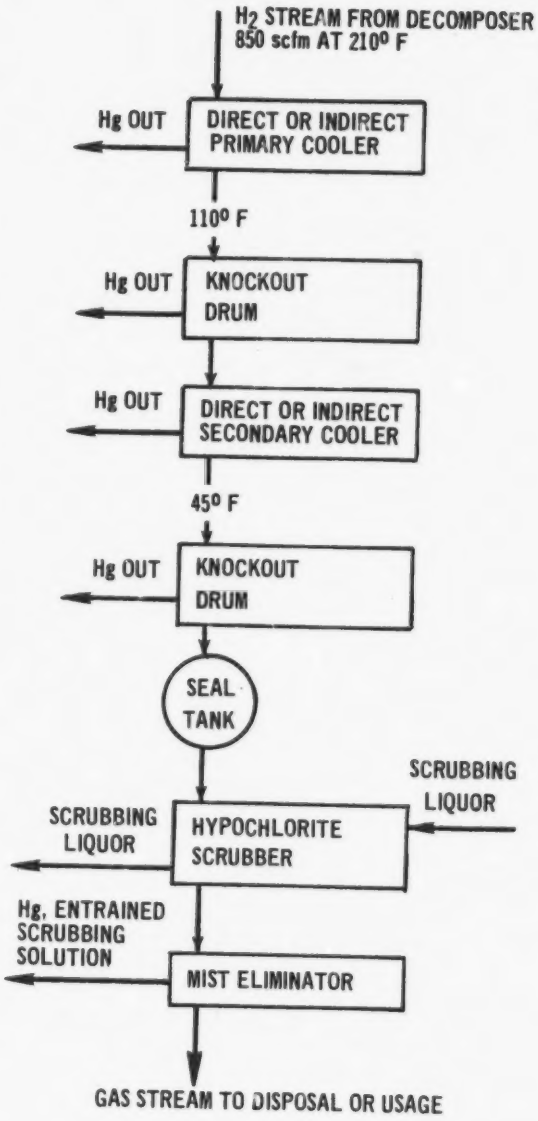

Figure 3-18. Hypochlorite scrubbing system.

3.2.4.4 Treated Activated Carbon

Control systems containing either sulfur- or iodine-impregnated activated carbon are being utilized by several mercury-cell chlor-alkali plants for reduction of the mercury concentration in the hydrogen stream. In

these systems, the mercury vapor is adsorbed by the carbon and chemically reacts with the sulfur or iodine to form mercury compounds. If properly designed, this technique can reduce the mercury concentration in the hydrogen stream to 5 to 10 micrograms per cubic meter.[23] The hydrogen at the inlet to the treated activated carbon bed usually has had 90 percent of its mercury content removed by primary and secondary cooling followed by efficient mist elimination. Figure 3-19 is a schematic flow diagram for a typical hydrogen stream employing treated activated carbon. The treated activated carbon can adsorb from 10 to 20 percent of its weight in mercury before it requires replacement. Destructive distillation of saturated activated carbon in retorts appears practical for recovering the adsorbed mercury.

This technique should also be applicable to mercury removal from the end-box ventilation system.

3.2.4.5 Molecular Sieve

The molecular sieve control technique utilizes a sieve-adsorbent blend to adsorb the mercury contained in gas streams. A molecular sieve system currently available for treatment of the hydrogen gas stream of chlor-alkali plants is the PuraSiv–Hg System. The designer of this system guarantees a reduction of the mercury concentration level to 0.50 milligram per cubic meter.[36] This concentration corresponds to a hydrogen stream emission of 0.04 pound of mercury per 100 tons of daily chlorine capacity.

Figure 3-20 illustrates a simplified PuraSiv–Hg System for the hydrogen stream of a mercury-cell chlor-alkali plant.[36] Hydrogen, laden with mercury vapor and mist, is passed through a secondary cooler and a mist eliminator. The gas stream then passes through one of two adsorption beds, both of which contain a proprietary sieve-adsorbent blend. Eighty to 90 percent of the treated hydrogen gas, containing 0.50 milligram per cubic meter mercury or less, is vented to the atmosphere, combusted in a burner system, or used in a subsequent production operation.[36] The remainder of the controlled hydrogen

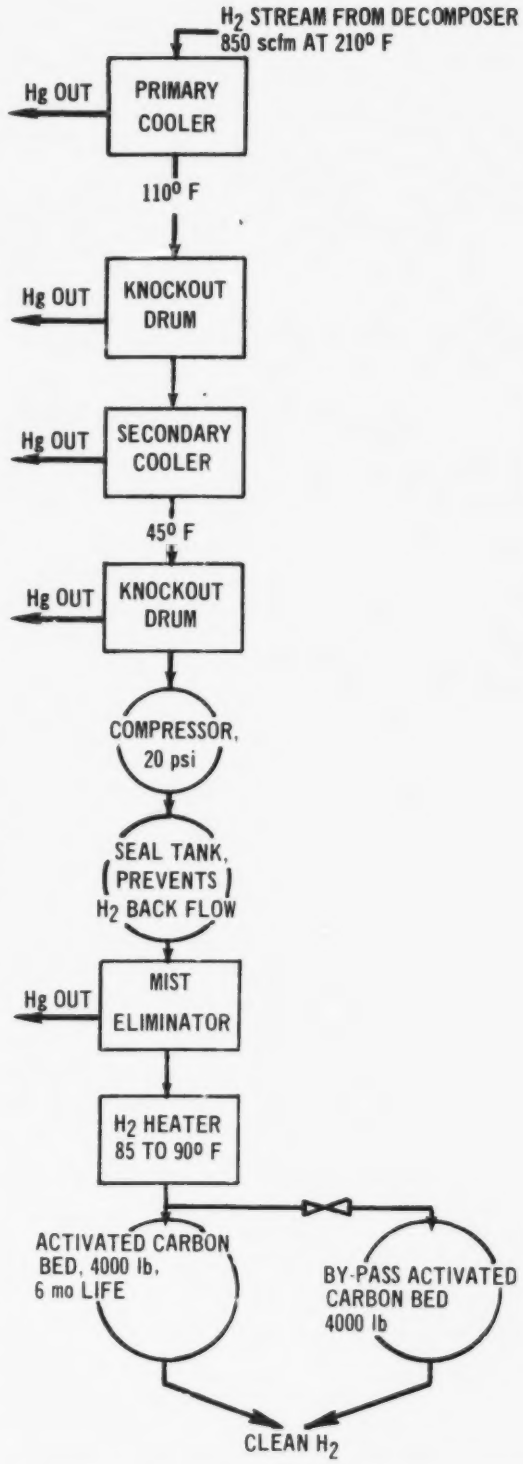

Figure 3-19. Activated carbon bed system.

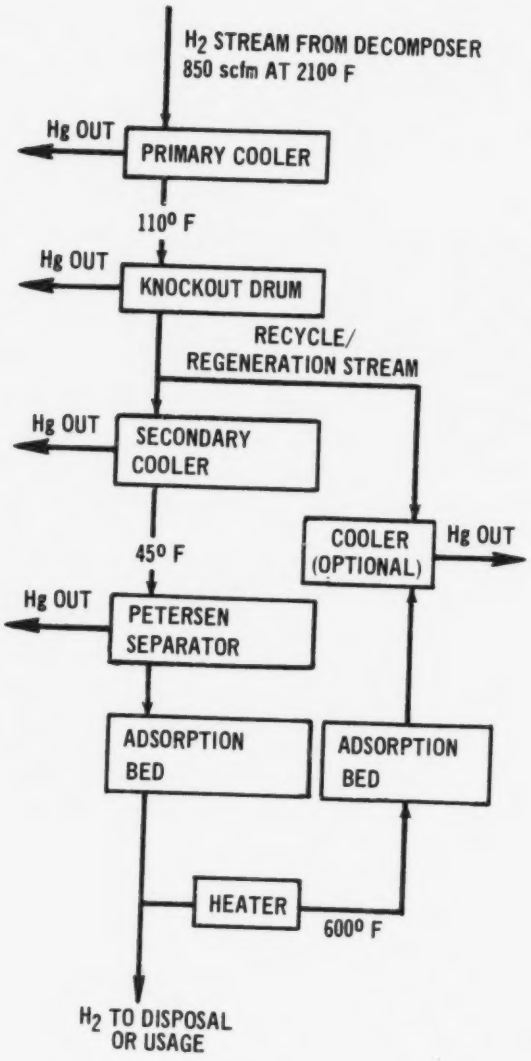

Figure 3-20. Process flow sheet for a two-bed molecular sieve system.

stream is heated to 600°F and used as a recycle-regeneration stream for removing entrapped mercury for the second adsorber bed. After passing through the second adsorption bed, this gas stream, with its high mercury concentration, passes through a cooler and is combined with the incoming mercury-laden hydrogen stream from the primary cooling section. The two adsorption beds alternate in function; while one bed is removing mercury from the gas stream, the other bed is being regenerated.

Since the mercury adsorption characteristics of the end-box ventilation gas stream are similar to those of the hydrogen stream, the molecular sieve system has potential application for the control of mercury emission from the end-box ventilation system. Special materials of construction would be required in most situations because of the more corrosive nature, due to chlorine and mercury salt contaminants, of this gas stream.

3.2.4.6 Housekeeping Practices

The following housekeeping practices for minimizing the various mercury emissions within the cell room are recommended.[40] Adherence to these recommended practices will result in a sizeable reduction of the mercury vapor concentration in the ventilation effluent from the cell room.

1. Chlorine cells and end-box covers should be installed, operated, and maintained in a manner to minimize leakage of mercury and mercury-contaminated materials.
2. Daily inspection should be made by operating personnel to detect leaks, and immediate steps to stop the leaks should be taken.
3. High housekeeping standards should be enforced, and any spills of mercury should be promptly cleaned up either mechanically or chemically or by other appropriate means. Each cell room facility should have available and should employ a well-defined procedure for handling these situations.
4. Floor seams should be smoothed over to minimize depressions and to facilitate washing down the floors.

5. All floors should be maintained in good condition, free of cracking and spalling, and should be regularly inspected, cleaned, and, to the extent practical, chemically decontaminated.
6. Gaskets on denuders and hydrogen piping should be maintained in good condition. Daily inspection should be made to detect hydrogen leaks, and prompt corrective action should be taken. Covers on decomposers, end-boxes, and mercury pump tanks should be well maintained and kept closed at all times except when operation requires opening.
7. Precautions should be taken to avoid all mercury spills when changing graphite grids or balls in horizontal decomposers or graphite packing in vertical decomposers. Mercury-contaminated graphite should be stored in closed containers or under water or chemically treated solutions until it is processed for reuse or disposed.
8. Where submerged pumps are used for recycling mercury from the decomposer to the inlet of the chlorine cell, the mercury should be covered with an aqueous layer maintained at a temperature below its boiling point.
9. Each submerged pump should have a vapor outlet with a connection to the end-box ventilation system. The connection should be under a slight negative pressure so that all vapors flow into the end-box ventilation system.
10. Unless vapor-tight covers are provided, end-boxes of both inlet and outlet ends of chlorine cells should be maintained under an aqueous layer maintained at a temperature below its boiling point.
11. End-boxes of cells should either be maintained under a negative pressure by a ventilation system or be equipped with fixed covers which are leak tight. The ventilation system or end-box covers should be maintained in good condition.
12. Any drips from hydrogen seal pots and compressor seals should be collected and confined for processing to remove mercury, and these drips should not be allowed to run on the floor or in open trenches.
13. Solids and liquids collected from back-flushing the filter used for alkali metal hydroxide should be collected in an enclosed system.
14. Impure amalgam removed from cells and mercury recovered from process systems should be stored in an enclosed system.
15. Brine should not be purged to the cell room floor. Headers or trenches should be provided when it is necessary to purge brine from the process. Purged brine should be returned to the system or sent to a treating system for mercury removal.
16. A portable tank should be used to collect any mercury spills during maintenance procedures.
17. Good maintenance practice should be followed when cleaning chlorine cells. During cleaning, all cells should have any mercury surface covered continuously with an aqueous medium. When the cells are disassembled for overhaul maintenance, the bed plate should be either decontaminated chemically or thoroughly flushed with water.
18. Brine, alkali metal hydroxide, and water-wash process lines and pumps should be maintained in good condition, and leaks should be minimized. Leaks should be corrected promptly, and in the interim, the leaks should be collected in suitable containers rather than allowed to spill on floor areas.

3.2.5 Control Costs

Costs in this section are developed for a model plant of 100 tons per day of chlorine capacity. Many actual equipment costs were received by communication with several mercury-cell chlor-alkali plant operators and with various vendors of equipment and proprietary processes. The various capital

costs are itemized and documented in the Appendix, Section A.3, for specific systems of control equipment. This section will classify control systems and illustrate their respective capital and annual costs. The methods of cost estimation are presented in Section A.2.

3.2.5.1 Conversion to Diaphragm-Cell Chlor-Alkali Plant

The technology and problems involved in converting a mercury-cell chlor-alkali plant to a diaphragm-cell chlor-alkali plant are discussed in Section 3.2.3. A recent paper indicates that the conversion cost will be approximately $12,000 per daily ton of chlorine capacity;[41] thus, about $1,200,000 of capital investment would be required for the conversion of a plant producing 100 tons of chlorine per day. Table A-9 in the Appendix presents the conversion cost for a 100-ton-per-day plant. Estimates range from $3,700,000 to $8,000,000 if the 11 percent diaphragm cell sodium hydroxide is upgraded and purified to the quality of the caustic produced by the mercury cell.

3.2.5.2 Cooling and Mist Elimination

A control system utilizing primary cooling and partial mist elimination by means of a knockout drum is considered to be the minimum existing technology practiced at most domestic plants. This control system is applicable to both the hydrogen and the end-box ventilation streams. The capital cost and annual operating cost for this system are $49,000 and $15,000, respectively, if applied to both streams. Expected emissions from the combined hydrogen and end-box streams when treated by this system are listed in Table A-8, which contains emission and cost data for several control systems. A complete breakdown of each system's cost is presented in Table A-7.

The costs of most heat exchanger equipment for the hydrogen system were calculated on the basis of stainless steel as the construction material. Costs were adjusted for the end-box ventilation system to reflect the higher cost of titanium and titanium-clad materials necessitated by the more corrosive nature of this stream. Several metals are listed below in order of increasing resistance to the corrosion effects of the end-box ventilation gases:

1. Mild steel.
2. Alloy 316 stainless steel.
3. Titanium-clad steel.
4. Solid titanium.

It has been estimated that the addition of a secondary cooler, a knockout drum, and a mist elimination device to the base system, as illustrated in Figure 3-16, could reduce the combined emission of the hydrogen and end-box streams to 3.4 pounds per day for a 100-ton-per-day plant. The capital and annual operating cost for this system would be $202,000 and $60,000 for the treatment of both process streams.

3.2.5.3 Chemical Scrubbing

Figure 3-17 illustrates a depleted brine scrubbing system, which is applicable to both the hydrogen and the end-box ventilation stream. Table A-7 in the Appendix gives a breakdown of the estimated costs for a plant producing 100 tons per day of chlorine. Estimated emissions and total system costs for plants of 100, 250, and 500 tons per day are listed in Table A-8. System costs range from $160,000 to $350,000 for a 100-ton-per-day plant. Annual operating costs are estimated to be $48,000 to $105,000.

A diagram of a hypochlorite scrubbing system, which is applicable to both the hydrogen and end-box stream, is presented in Figure 3-18. Estimated emissions for a 100-ton-per-day plant are 0.2 to 0.8 pound per day for both process streams. Capital cost and annual operating costs for such a system would be approximately $226,000* and $68,000. More complete data on capital cost and expected emissions are provided in Tables A-6, A-7, and A-8.

3.2.5.4 Treated Activated Carbon

Figure 3-19 illustrates a control system that has been successfully employed on all or part of the hydrogen streams from several domestic mercury-cell chlor-alkali plants. Table A-7 in the Appendix lists the breakdown for the costs of the various pieces of equipment required. The total capital investment for a plant producing 100 tons of

*Does not include licensing fee.

chlorine per day is estimated to be $279,000; the operating cost is estimated at $83,000 per year. These costs are for the application of treated activated carbon on both the hydrogen and end-box ventilation gas streams.

3.2.5.5 Molecular Sieve

The molecular sieve system, which is illustrated in Figure 3-20, can be applied to both the hydrogen and end-box stream. By treating both streams, it is estimated that total emissions could be reduced to 0.08 pound of mercury per 100 tons of daily chlorine capacity. Total capital investment for a 100-ton-per-day plant is estimated to be $349,000. Annual operating cost is expected to be $105,000.

3.2.5.6 Summary

The approximate costs of mercury control systems and the associated pieces of equipment have been developed and are presented in this section and Section A.3. Since a 100-ton-per-day chlorine capacity has been used for all cost estimations, the following equation is presented for developing cost estimates for other capacity plants.

$$C_A = C_{100} \left(\frac{P_A}{100 \text{ tons/day}} \right)^{0.6}$$

where: C_A = applicable control cost; equipment, capital, or operating

C_{100} = control cost for 100-ton-per-day plant; equipment, capitol, or operating

P_A = applicable plant chlorine capacity in tons per day

Table 3-6 is a summary of control systems costs. As a basis for comparison of the cost of various control systems, the capital requirement for the construction of a grass-roots mercury-cell chlor-alkali plant is utilized as a common denominator. An estimate of this capital requirement is $100,000 for each daily ton of chlorine capacity, or $10,000,000 for a 100-ton-per-day plant.

All costs that have been discussed thus far have not taken one possible contingency into consideration. This contingency is the additional cost of procuring proprietary equipment, systems, or associated engineering.

3.2.6 Development of New Technology

3.2.6.1 Ion Exchange Process

A Japanese ion exchange process has been licensed for domestic use for the removal of mercury from both air and water. The designer claims that this process will reduce the mercury concentration in a gas stream to 10 micrograms per cubic meter.[42] There are no known domestic applications of this process on either the hydrogen or the end-box ventilation streams of mercury-cell chlor-alkali plants.

3.2.6.2 Sulfuric Acid Scrubber

As previously discussed in Section 3.1.5.2, a foreign nonferrous smelting operation has employed a sulfuric acid scrubbing system to remove mercury and selenium from its sulfur dioxide gas stream.[29,30] Current information indicates that this method has not been employed in any mercury-cell chlor-alkali plant in the world for control of mercury emissions from the hydrogen and end-box ventilation gas streams.

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*Office of Air Programs, Stationary Source Pollution Control Programs, Standards Development Implementation Division.

Table 3-6. SUMMARY OF CHLOR-ALKALI CONTROL COSTS AND EXPECTED EMISSIONS FOR COMBINED HYDROGEN AND END-BOX VENTILATION STREAMS FOR A 100-TON/DAY FACILITY

Control system	Capital cost, $	Annual operating cost, $	Annual operating cost as % of grass roots plant cost	Estimated emissions from combined H_2 and end-box streams, lb/day
Diaphragm cell conversion (Section 3.2.5.1, Table A-9)	3,700,000 to 8,000,000	1,000,000 to 2,400,000	11.0 to 24.0	0.0
Cooling and mist elimination (Section 3.2.5.2, Figure 3-16, and Tables A-7 and A-8)	201,000	60,000	0.6	3.4
Chemical scrubbing (Section 3.2.5.3, Figures 3-17 and 3-18, and Tables A-7 and A-8)	159,000 to 349,000	48,000 to 105,000	0.5 to 1.1	0.02 to 0.6
Treated activated carbon (Section 3.2.5.4, Figure 3-19, and Tables A-7 and A-8)	278,000	83,000	0.8	0.3
Molecular sieve (Section 3.2.5.5, Figure 3-20, and Tables A-7 and A-8)	349,000	105,000	1.0	0.1

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APPENDIX

A.1 CALCULATION OF EQUILIBRIUM CONCENTRATION OF MERCURY VAPOR IN A GAS AND RESULTANT LOSSES IN PROCESS STREAMS OR IN VENTILATION AIR

The weight of mercury, W, contained per unit volume of gas at equilibrium can be calculated from the ideal gas law, PV = nRT, or, in this instance,

$$W/V = PM/RT$$

where: W = weight of Hg, mg
V = volume of gas, m^3
P = equilibrium vapor pressure of Hg, mm Hg at temperature of saturated gas stream
M = molecular weight of Hg = 200.6 x 10^3 mg/mole
T = absolute temperature, °K
R = gas constant = 0.06237 (m^3) (mm Hg)/(°K)(mole)

thus W/V (mg/m^3) = 1.09 x 10^4 x P at standard conditions of 70°F and 29.92 inches of Hg.

The equilibrium concentration of mercury is plotted as a function of Fahrenheit temperature in Figure A-1. These values can be used to estimate the mercury vapor losses to be expected in a mercury-saturated gas stream as a function of the condenser operating temperature. For this calculation, it is assumed that approximately 300 cubic meters of hydrogen is produced and vented per ton of chlorine produced. Some representative calculated values are given in Table A-1. The mercury vapor pressure data used were taken from the *Handbook of Chemistry and Physics.*[1] The equilibrium concentration of mercury can also be used to estimate the order of magnitude of mercury emissions that might be associated with the venting of the end-box ventilation air. In this application the mercury in the end-box is covered by an aqueous layer to prevent rapid evaporation of the mercury; hence the end-box air may not

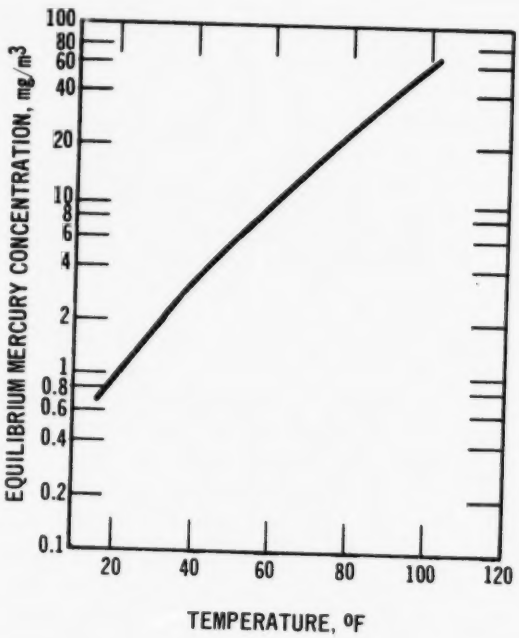

Figure A-1. Equilibrium concentration of mercury vapor in air as a function of temperature.

be saturated with mercury vapor. If, however, a mean air temperature, a degree of saturation, and a ventilation air flow rate are assumed, the mercury emission rate, E, in pounds per day can be calculated from the equation:

$$E = 0.09 \times 10^{-6} \times V \times C,$$

where: E = mercury emission rate, lb/day
V = air flow rate, cfm
C = mercury concentration, $\mu g/m^3$

For example, assuming an end-box ventilation flow rate of 1000 standard cubic feet per minute, a mean air temperature of 100°F (equilibrium concentration of mercury = 57 milligrams per cubic meter), and a 50 percent saturation of the air, the mercury emission rate would be:

Table A-1. MERCURY VAPOR LOSSES IN A HYDROGEN STREAM[a]

Condenser temperature		Mercury concentration, mg/m^3 [b]	Mercury loss/ton Cl_2		Mercury loss/day for 100-T plant	
°C	°F		g	lb	g	lb
40	104	67	20	0.040	2000	4.4
30	86	30	9.1	0.020	910	2.0
25	77	23	6.8	0.015	680	1.5
20	68	13	4.0	0.008	400	0.80
14	57	7.8	2.4	0.005	240	0.51
4	39	3.0	0.90	0.002	90	0.20
0	32	2.0	0.60	0.0014	60	0.14
-10	14	0.66	0.20	0.0004	20	0.04

[a]Assume $300m^3$ of hydrogen per ton of chlorine.
[b]At 70°F and 29.92 in. Hg.

$E = 0.09 \times 10^{-6} \times 1000 \times (0.50 \times 57{,}000) = 2.6$ lb/day.

The calculation can be repeated for any number of assumed conditions. When the results obtained for an arbitrary series of conditions are plotted as emission rate versus ventilation air flow rate, a family of straight lines is obtained, as shown in Figure A-2. Actual ventilation flow rates used in the operations that were surveyed ranged from 400 to 2000 cubic feet per minute. As documented in the body of this report, mercury emissions in the end-box ventilation air have been noted to vary from 2 to 15 pounds per day for each 100 tons per day of chlorine produced. The temperatures of untreated end-box ventilation air streams ranged from approximately 100° to 160°F and had mercury vapor saturations of about 10 to 80 percent.

The above equation may also be used to calculate mercury emissions in building ventilation air. The mercury vapor concentration in this case is very low. If the actual value is not known, the maximum emission rate may be estimated by assuming the concentration to be equal to the Threshold Limit Value; that is, 50 micrograms per cubic meter. The volumes encountered in the cell room ventilation air ranged from 100,000 to 1,000,000 cubic feet per minute. A few calculated emission rates for arbitrary air flow rates in these ranges are given in Table A-2.

A.2 METHODS OF ESTIMATING CONTROL COSTS

A.2.1 Equipment Costs

The equipment costs used in this document are the free-on-board (f.o.b.) charges for either a specific piece or system of control equipment. Unless otherwise indicated, the equipment costs are based on the use of stainless steel as the construction material. When more corrosion-resistant materials such as titanium or titanium-clad steel are required, the equipment costs are estimated to be 25

Table A-2. CALCULATED MERCURY VAPOR EMISSION RATES IN CELL ROOM VENTILATION AIR[a]

Air flow, cfm	Mercury emission rate, lb/day
1,000	0.0045
2,500	0.011
5,000	0.023
10,000	0.045
100,000	0.45
250,000	1.15
500,000	2.3
1,000,000	4.5

[a]Assume a mercury concentration of 50 $\mu g/m^3$.

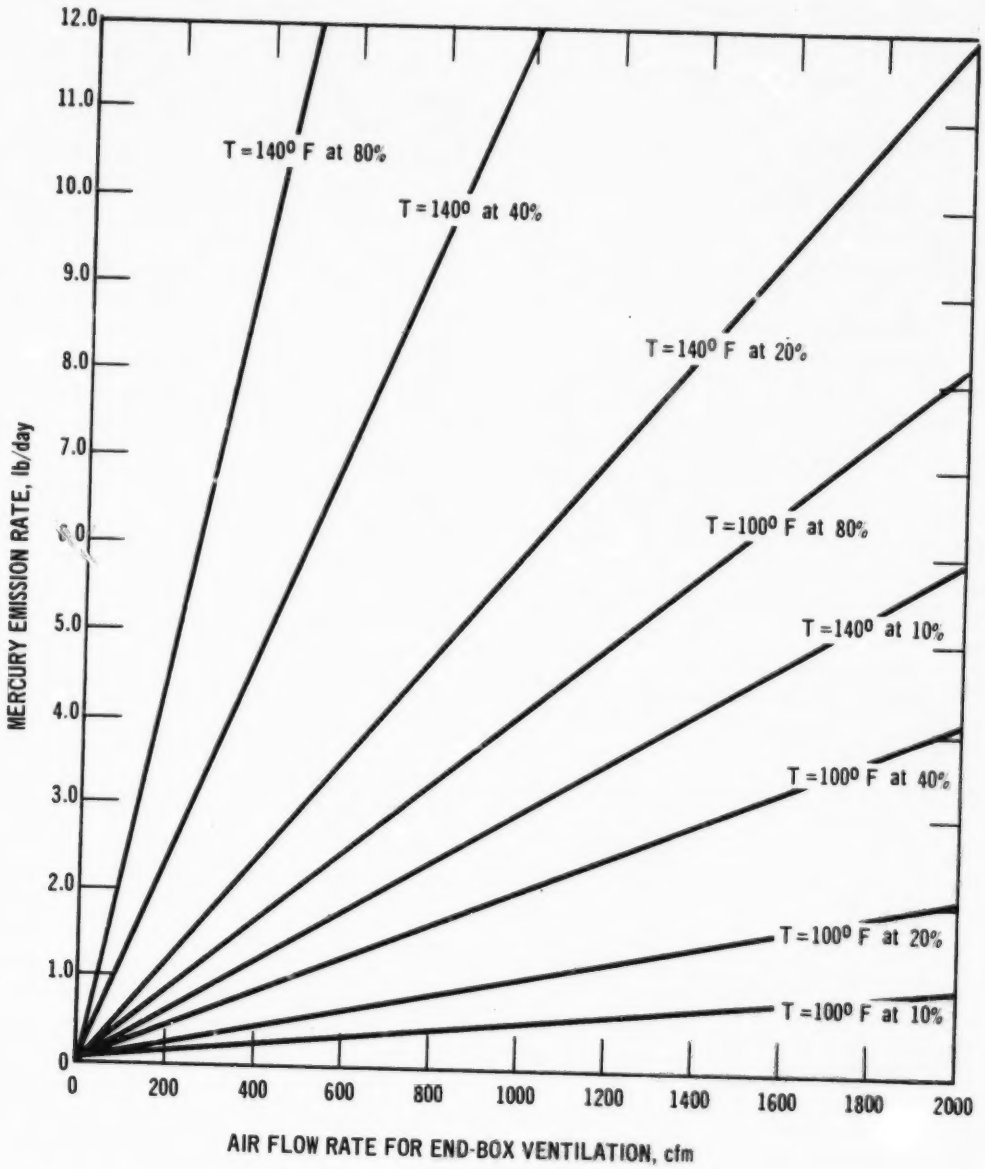

Figure A-2. Mercury emission rate as a function of end-box ventilation air flow rate at arbitrary conditions of air temperature (T) and percentage saturation (%).

percent greater than the cost based on stainless steel construction.

All equipment costs were obtained through communications with vendors and users of said equipment. In most situations, averages of f.o.b. estimates from vendors and users were selected. These f.o.b. estimates serve as a basis for the calculation of other costs, such as the fixed-capital requirement and the annual operating cost.

A.2.2 Fixed-Capital Requirement

The fixed-capital requirement for a piece or system of control equipment includes the following:[2]

1. Purchase price.
2. Cost of installation.
3. Instrumentation and control.
4. Piping.
5. Electrical equipment and materials.
6. Engineering and supervision.
7. Construction expenses.
8. Contingency.

In this list, the cost of installation includes costs for labor, foundations, supports, platforms, construction, and other items directly related to the erection of the purchased equipment.[2]

In order to estimate the fixed-capital requirement, the following assumptions were made. All cost estimates are presented as a percentage of the equipment cost based on stainless steel construction.[3]

1. Additional buildings, service facilities, and site preparation are unnecessary.
2. The cost of installation equals the equipment cost.
3. Process piping, electrical equipment, and instrumentation are in accordance with the minimum requirements. The combined cost of these items is equal to 40 percent of the equipment cost.
4. Engineering expenses are approximately 30 percent of the equipment cost.
5. Construction overhead adds an amount equal to 20 percent of the equipment cost. This estimate is for applications involving nonexplosive gas streams. For control systems involving the hydrogen gas stream of chlor-alkali plants, the construction overhead is 80 percent of the equipment purchase cost.
6. Plant maintenance personnel provide most of the construction force.
7. An amount equal to 30 percent of the equipment cost is added to account for contingencies.

If the preceding components of the fixed-capital requirement are added to the equipment purchase cost, estimates obtained of the fixed-capital requirement range from 320 to 380 percent of the equipment cost. These estimates are for nonexplosive and explosive gas streams, respectively.

For applications requiring the use of titanium or titanium-clad steel construction material, the estimated fixed-capital requirement is increased by an amount equal to 25 percent of the stainless steel equipment cost. The estimated fixed-capital requirement for titanium equipment would therefore range from 345 to 405 percent of the equipment purchase cost.

The fixed-capital estimation method presented above may tend to result in high estimates because the equipment costs are based on stainless steel construction.[4] The accuracy of the estimation method is approximately 30 percent.

The costs presented in this document were fixed on January 1, 1972. Costs for later dates can be estimated by using appropriate indices.

A.2.3 Annual Operating Costs

The annual operating costs are estimated to be 30 percent of the estimated capital requirement.[3] This estimating procedure is based on accepted standard practice in the chemical process industry and should provide an ample allowance. The estimate includes allowances for labor and supervision, maintenance, payroll overhead, operating supplies, indirect costs, and capital charges at 18 percent per year.[3]

A.3 CONTROL EQUIPMENT COSTS AND EFFICIENCIES

Control efficiencies and equipment costs are itemized in Tables A-3 through A-9.

A.4 GEOGRAPHIC LOCATION OF CHLOR-ALKALI PLANTS THAT USE MERCURY CELLS

The chlor-alkali plants that use mercury

cells are listed in Table A-10. A map showing these locations is presented in Figure A-3.

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Table A-3. ESTIMATED COLLECTION EFFICIENCIES OF CONTROL EQUIPMENT

Equipment	Estimated collection efficiency
Heat exchanger and knockout drum	80% (particulate removal)
Mist eliminator	86% (particulate removal)
Venturi scrubber	95% (particulate removal)
Depleted brine scrubbing system (system A)	Exit concentration of 85 $\mu g/m^3$ (particulate and vapor removal)[a]
Depleted brine scrubbing system (system B)	Exit concentration of 4.4 mg/m^3 (particulate and vapor removal)[a]
Hypochlorite scrubbing system	95 to 99% (particulate and vapor removal)
Activated carbon system	90% (vapor removal)
Molecular sieve system	Exit concentration of 0.5 mg/m^3 (vapor removal)[a]

[a]Concentrations will be reduced to this level, regardless of loadings, according to the equipment manufacturers.

Table A-4. EQUIPMENT COSTS FOR PRIMARY MERCURY EXTRACTION FACILITIES AT 100-TON/DAY CAPACITY

Equipment	Purchase cost (stainless steel), $	Purchase cost (titanium), $	Capital cost (titanium), $
Secondary cooler (indirect)	8,300	10,400	28,600
Chiller	14,000	–	44,900
Knockout drum	600	750	2,100
Mist eliminator	9,500	11,900	32,800
Venturi scrubber	3,700	4,600	12,800
Carbon bed system	15,500	19,400	53,500

Table A-5. SYSTEM COSTS FOR PRIMARY MERCURY EXTRACTION FACILITIES WITH 100-TON/DAY CAPACITY

System	Equipment	Capital costs, $	Emissions, lb/day
Cooling and partial mist elimination	Secondary cooler (indirect)	28,600	5.8
	Chiller	44,900	
	Knockout drum	2,100	
Total		75,600	
Cooling and mist elimination	Secondary cooler (indirect)	28,600	1.7
	Chiller	44,900	
	Knockout drum	2,100	
	Mist eliminator	32,800	
Total		108,400	
Wet scrubbing and carbon bed	Venturi scrubber	12,800	1.8
	Carbon bed system	53,500	
Total		66,300	
Cooling and wet scrubbing	Secondary cooler (indirect)	28,600	1.7
	Chiller	44,900	
	Venturi scrubber	12,800	
Total		86,300	

Table A-6. EQUIPMENT COSTS FOR MERCURY-CELL CHLOR-ALKALI PLANTS WITH 100-TON/DAY CAPACITY

Equipment	Purchase cost (H_2 stream), $	Capital cost (H_2 stream), $	Purchase cost (end-box stream), $	Capital cost (end-box stream), $
Primary cooler (indirect)	5,700	21,700	7,100	19,700
Secondary cooler (indirect)	5,700	21,700	7,100	19,700
Chiller	9,600	30,700	9,600	30,700
Seal tanks	1,200	4,600	–	–
Knockout drum	400	1,500	500	1,400
Blower	400	1,500	500	1,400
Mist eliminator	6,500	24,700	8,300	22,400
Petersen separator	3,800	14,400	4,800	13,100
Depleted-brine scrubbing system (system A)	20,800	79,000	26,000	71,800
Depleted-brine scrubbing system (system B)	15,500	58,900	19,400	53,500
Hypochlorite scrubbing system	6,000	22,800	7,500	20,700
Carbon bed system	10,600	40,300	13,300	36,600
Molecular sieve system	23,000	87,400	28,800	79,400

Table A-7. CONTROL SYSTEM COSTS FOR CHLOR-ALKALI PLANTS TREATING HYDROGEN AND END-BOX VENTILATION STREAMS BASED ON 100-TON/DAY CAPACITY

System	Equipment	Capital cost (H_2 stream), $	Capital cost (end-box stream), $
Partial cooling and mist elimination (see Figure 3-14 for basic system)	Primary cooler	21,700	19,700
	Knockout drum	1,500	1,400
	Seal tanks	4,600	–
Total		27,800	21,100
Cooling and mist elimination (Figure 3-16)	Base system	27,800	21,100
	Secondary cooler	21,700	19,700
	Chiller	30,700	30,700
	Knockout drum	1,500	1,400
	Mist eliminator	24,700	22,400
Total		106,400	95,300
Cooling, mist elimination, and chemical scrubbing (Figure 3-17)	Base system	27,800	21,100
	Secondary cooler	21,700	19,700
	Chiller	30,700	30,700
	Mist eliminator	24,700	22,400
	Depleted-brine scrubbing system (system A)	79,000	71,800
Total		183,900	165,700
Cooling, mist elimination, and chemical scrubbing (Figure 3-17[a])	Mist eliminator	24,700	22,400
	Depleted-brine scrubbing system (system B[b])	58,900	53,500
Total		83,600	75,900
Cooling, mist elimination, and chemical scrubbing (Figure 3-18)	Base system	27,800	21,100
	Secondary cooler	21,700	19,700
	Chiller	30,700	30,700
	Knockout drum	1,500	1,400
	Hypochlorite scrubbing system[c]	22,800	20,700
	Petersen separator	14,400	13,100
Total		118,900	106,700
Cooling, mist elimination, and activated carbon (Figure 3-19)	Base system	27,800	21,100
	Secondary cooler	21,700	19,700
	Chiller	30,700	30,700
	Knockout drum	1,500	1,400
	Mist eliminator	24,700	22,400
	Carbon bed	40,300	36,600
Total		146,700	131,900
Cooling, mist elimination, and molecular sieve (Figure 3-20)	Base system	27,800	21,100
	Secondary cooler	21,700	19,700
	Chiller	30,700	30,700
	Blower	1,500	1,400
	Petersen separator	14,400	13,100
	Molecular sieve system[d]	87,400	79,400
Total		183,500	165,400

[a]System does not include alkaline scrubber.
[b]Includes cost of primary cooler and compressor.
[c]Does not include licensing fee.
[d]Includes preheater, two absorption beds, and 600°F H_2 heater.

Table A-8. CONTROL SYSTEM COSTS AND EMISSION RATES FOR COMBINED HYDROGEN AND END-BOX VENTILATION STREAMS

System	Production rate, tons/day	Capital cost, $	Emissions, lb/day
Partial cooling and mist elimination (see Figure 3-14 for base system)	100	49,000	99.6
	250	85,000	249.0
	500	129,000	498.0
Cooling and mist elimination (Figure 3-16)	100	202,000	3.4
	250	350,000	8.5
	500	531,000	17.0
Cooling, mist elimination, and chemical scrubbing (Figure 3-17) (system A)	100	350,000	0.02
	250	607,000	0.05
	500	919,000	0.10
Cooling, mist elimination, and chemical scrubbing (Figure 3-17) (system B)[a]	100	160,000	0.6
	250	277,000	1.5
	500	420,000	3.0
Cooling, mist elimination, and chemical scrubbing (Figure 3-18)[b]	100	226,000	0.2
	250	392,000	0.5
	500	594,000	1.0
Cooling, mist elimination, and activated carbon (Figure 3-19)	100	279,000	0.28
	250	483,000	0.70
	500	733,000	1.40
Cooling, mist elimination, and molecular sieve (Figure 3-20)	100	349,000	0.08
	250	605,000	0.20
	500	917,000	0.40

[a]System does not include alkaline scrubber.
[b]Does not include licensing fee.

Table A-9. CAPITAL INVESTMENT REQUIRED TO CONVERT FROM MERCURY TO DIAPHRAGM CELLS AT PLANT WITH 100-TON/DAY CAPACITY[4,a]

	Capital investment, $
Site preparation including grading, sewers, fire protection, and road and fence changes	230,000
New building for cells	500,000
Cells including headers, anodes, bus, cell rebuilding facilities, and rectifier changes	1,200,000
Weak-liquor handling and storage	190,000
Weak-liquor evaporation plant including salt handling	1,600,000
New boiler to supply steam for weak-liquor evaportation plant	800,000
Caustic purification	1,693,000
Engineering and construction supervision	1,552,000
Contingency	625,000
Dismantling of old mercury cell facility	150,000
Credit for mercury recovered	(540,000)
Total cost	8,000,000

[a]Assumptions: (1) Diaphragm cell would be installed in a new building in order to permit continued operation of old plant until diaphragm cells are on-line; (2) Diaphragm cell caustic would be purified to give quality equivalent to mercury cell liquor.

Table A-10. MERCURY-CELL CHLOR-ALKALI PLANTS IN THE UNITED STATES

State and city	Producer	Year built[a]	Cell type
Alabama			
Le Moyne	Stauffer Chemical Co.	1965	De Nora 22 x 5
McIntosh	Olin Corp.	1952	Olin E8
Mobile	Diamond Shamrock Chemical Co.	1964	De Nora
Muscle Shoals	Diamond Shamrock Chemical Co.	1952	De Nora 24 x 2M
Delaware			
Delaware City	Diamond Shamrock Chemical Co.	1965	De Nora 18 x 4
Georgia			
Augusta	Olin Corp.	1965	Olin E11F
Brunswick	Allied Chemical Corp.	1957	Solvay V-100
Illinois			
East St. Louis	Monsanto Co.	1962	De Nora 18 x 6
Kentucky			
Calvert City	B.F. Goodrich Chemical Corp.	1966	De Nora 24H5
Calvert City	Pennwalt Corp.	1967	Uhde 30 m^2
Louisiana			
Lake Charles	PPG Industries, Inc.	1969	De Nora 48H5
St. Gabrile	Stauffer Chemical Co.	1970	Uhde 30 m^2
Geismar	BASF Wyandotte Corp.	1964	Uhde 30 m^2
Maine			
Orrington	Sobin Chlor-Alkali, Inc.	1967	De Nora 24H5
New York			
Niagara Falls	Hooker Chemical Corp.	1961	Uhde 20 m^2
Niagara Falls	Olin Corp.	1960	Olin E11F
Syracuse	Allied Chemical Corp.	1946	Solvay Process SD 12
Syracuse	Allied Chemical Corp.	1953	Solvay S60
North Carolina			
Pisgah Forest	Olin, Ecusta Operations	1947	Sorensen
Acme	Allied Chemical Corp.	1963	Solvay V-200
Ohio			
Ashtabula	Detrex Chemical Industries, Inc.	1963	Olin E11F
Tennessee			
Charleston	Olin Corp.	1962	Olin E11F, E812
Texas			
Dear Park	Diamond Shamrock Chemical Co.	1938	De Nora 18 SGL
Point Comfort	Aluminum Co. of America	1966	De Nora 24 x 5
Washington			
Bellingham	Georgia-Pacific Corp.	1965	De Nora 18 x 4
Longview	Weyerhaeuser Co.	1967	De Nora 14 TGL & 24H5
West Virginia			
Moundsville	Allied Chemical Corp	1953	Solvay S60
New Martinsville	PPG industries, inc.	1958	Uhde 20 m^2
Wisconsin			
Port Edwards	BASF Wyandotte Corp.	1967	De Nora 24H5

[a]Refers to year chlorine production started at location.

Figure A-3. Location of mercury-cell chlor-alkali plants in the United States.

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